GREEN CORROSION INHIBITORS FORMETALS AND ALLOYS: A COMPREHENSIVE REVIEW

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Corrosion inhibitors are widely used in many industries to reduce the corrosion of metals and alloys in contact with a corrosive environment. Most corrosion inhibitors are synthetic organic and inorganic chemicals, expensive, and very hazardous to the environment. Green inhibitors for protection of metals and alloys are of good alternative of synthetic chemical inhibitors. Natural occurring compounds are a good source of green corrosion inhibitors, where most of their extracts containing fictional groups along with hetero-atoms which are active in green compounds, assist in adsorption of these compounds on metal surfaces to form a barrier that protects the metallic surface and retards corrosion process. Development of green chemistry and green chemical technologies offers novel synthetic methods for the development new generation compounds which are considered as new green corrosion inhibitors. In this review briefly discuss the potential of various plants extract as a green corrosion inhibitor on metal surfaces like aluminium, mild steel, carbon steel, stainless steel, copper and zinc in corrosive environment by weight loss, potentiodynamic polarisation, gravimetric techniques. The inhibition efficiency has also been discussed in detail. This study will further support in the understanding of the mechanism of adsorption involved in inhibition and explore inhibition property of plant extract against metal deterioration.

Introduction:
Deterioration is a natural process where metals and alloys are reverting into a more stable state due to redox reaction with the surrounding environment. Corrosion is more expensive due to loss of metals and their resistance properties during uses. Due to lack of maintenance, the shutting down of installations, and severe failure of some structural skeletons, which in some cases may be hazardous to environment and cause human health issue.

The protection of metals by inhibitors is a most practical approach in various corrosive environments. Many organic and inorganic compounds have been used as inhibitors to protect metals from the attacking of corrosive agents. Generally, compounds that exert a significant effect on the extent of adsorption on the metallic surface and therefore may be used as effective inhibitors. The efficiency of these corrosion inhibitors is related to the presence of polar functions with S, O and N atoms in the molecule. The lone-electrons are usually regarded as the reaction point for the establishment of the adsorption phenomenon. The known hazardous effects of most synthetic organic inhibitors and restrictive environmental regulations have now made researchers to motivate to develop cheap and eco-friendly natural products as some inhibitors for metal protection.
Over the two decade, green inhibitors have been emphasizing the role of protecting the environment and human health in an economically beneficial aspect aiming at avoiding side effects and minimizing wastes generation. Generally, green corrosion inhibitors are biodegradable, ecologically acceptable and renewable.

Plant extracts are viewed as an incredibly rich source of naturally synthesized compounds that can be extracted by simple procedures with low cost and are biodegradable in nature. The use of these naturally occurring compounds such as extracted from various parts of plants as inhibitors have been widely reported by several investigators. Due to biodegradability and toxic less properties, several investigators worked on successful application of natural product as an inhibitor for mitigation of metallic corrosion in various environments.

To protect the metallic structures from corrosion, approaches such as isolating the structure from aggressive environments. Therefore, their use has been limited by environmental regulation authorities. The inhibitors still play a major role in metal corrosion prevention. Thus, green corrosion inhibitors are classified according to:

First, three types based on electrode function, namely, anodic inhibitors, cathodic inhibitors, and mixed inhibitors;

Second, the nature of the environment to organic and mineral acid inhibitors neutral or aqueous inhibitors, basic inhibitors, vapour-phase inhibitors and microbial inhibitors.

Most of these inhibitors isolated from natural products which act as are eco-friendly corrosion inhibitors and not create secondary problem on disposal site. This work will focus on green inhibitors which isolated from natural products.

Green inhibitors act when they are added in very trace amounts in corrosive environment. Plant extracts are deemed to be rich, naturally synthesized chemical compounds that directly affect the metal loss by adsorption of effective compounds on interface when added to environments through:

Changing the rate of redox reactions at interface;

Effecting the diffusion of corrosive ions interacting with scale which formed on surfaces;

Growing strong barrier on metal surface through adsorption of green inhibitors.

During the interaction of metal with environment, at anode metal is oxidised to form metal ions move into the environment and release electrons which an accept at cathodic site, there oxygen or hydrogen ions present as electron acceptor. Oxidation of metal can be minimized or controlled by retarding the oxidation or reduction reactions, or both (redox process). When inhibitors are added in the environment which adsorbed on the exposed metal surface and ultimately formed a strong protective film which checked interact with anodic and/or cathodic reaction process to decrease the oxidation and/or reduction reactions.

Generally, hetero-atom containing organic compounds are frequently used as corrosion inhibitors in various environments. However, due to low cost and easily available natural products an excellent alternative as good corrosion inhibitors in acidic, basic and aqueous environment. The natural occurring compounds will become more important in the coming period. As shown in present work, natural products have already been extensively applied as a substitutes of synthetic compounds as inhibitor for metal protection.

The eco-friendly natural products have been investigated as good corrosion inhibitors for metals and alloys. Table 1 shows the distribution of the research work published in the last decades on the various parts of plants extract used, which are presented in this review article. The majority of these studies have been focused on plant extracts. The main constituents of the various parts of plant extracts have been reported by many investigators. The research work performed in the last decades on various parts of plant extracts as corrosion inhibitors for metals and alloys in various environments (Table 1)

**Table 1:** Various parts of plant extract used as green corrosion inhibitor.

| Serial No. | Natural Products (extracts) | Uses (%) |
|------------|-----------------------------|----------|
| 1          | Leaves                      | 52       |
| 2          | Seeds                       | 16       |
| 3          | Barks                       | 14       |
Plant extracts as green corrosion inhibitors:
Several studies reported that the use of plants extract acts as good corrosion inhibitors for the control of metal losses in various corrosive environments (Table 2). Phytochemicals which present in the extracts have increasing interest in the field of sustainable materials. The evaluation of inhibition efficiency and durability of film formed on the metal surfaces are depend on nature of constituents of plants extracts.

Extraction methods of plant extract:
Many publications on the isolation and fractionation of different compounds such as plant extracts and purified compounds have appeared over the few decades. The method used must be carefully chosen according to the aim of the research. The selection of the extraction methods and medium are depending upon the nature of plants. In the extraction process, the selection of solvents playing important role because dissolution of phytochemicals in it. The yields of natural products purity are directly influenced by the nature of solvents.

The solvent extraction and the isolation procedures may have a significant impact on the extraction yield of constituents from the different part of plants. The extraction methods certainly have unique operating factors that affect the concentration and antioxidant activity of the extract, and need to be optimised. The extraction period, temperature, solvent percentage, sample number, choice of solvents are the main parameters which control extraction kinetics. Solubility of product is directly affected by the extraction period and temperature.

Conventional extraction processes are still generic due to their simplicity, efficiency, and more applicability. However, these traditional techniques have their own advantages and limitations. Solvent extraction results in a higher yield. The main drawbacks of this process are related to the use of conventional solvents such as methanol, ethanol, isopropanol, acetone, diethyl ether, and ethyl acetate, often mixed with various proportions of water for sample preparation, separation, detection, and the identification are of both environmental and economic concern.

Modern extraction methods are concern on reduced solvent methods, sorption-based methods, environmental applications, and on both solid and liquid samples. Supercritical fluid extraction (SFE), pressurised-liquid extraction (PLE), microwave-assisted extraction (MAE), solid-phase extraction (SPE), and solid-phase micro-extraction (SPME) offer important advantages over organic solvent technology, such as ecological friendly and ease of product fractionation. These methods became interesting due to their simplicity, shorter extraction period, and the release less amount of toxic pollutants by minimum organic solvent consumption. The most important advantage of SFE is the selective extraction of components or the fractionation of the total extracts.

Corrosion inhibition efficiency of plants extract:
In Table 2 more details the work published in the last decades on plant extracts as green corrosion inhibitors for aluminium and its alloys in various corrosive environments. Information on the extraction process and the main components of the extracts is also shown (Table 2).

As seen in Table 2, corrosion inhibition efficiency has been reported by many investigators for the aluminium and its alloys in various environments. The inhibition efficiency is calculated by weight loss and electrochemical methods. In most of the cases, it is observed that the various parts of plants extract act as good corrosion inhibitors and inhibit up to 95 to 98 percent. The inhibition efficiency of all studied extracts increased with increasing concentration, independently of their type, corrosive environment, and nature of metals.

The plant extracts studied as corrosion inhibitors were obtained from different parts of the plants, including fruits, leaves, stem and root bark, and seeds. The distribution of active phytochemicals in different parts of plants, such as such as leaves, stems, roots, and seed, shows qualitative and quantitative variations. Often different parts of the same plant contain completely different constituents. Many investigations have been performed evaluating the inhibition efficiency of different parts of the same plant extracted with the same solvent.

The change in the inhibition efficiency with temperature was found to be dependent on the corrosive environment in the case of Dendrocalamus brandisii leaf extract in ethanol. The investigators reported that the inhibition efficiency of the extract decreased with increasing temperature in HCl solutions, while remained almost constant in phosphoric...
The inhibition efficiency of all the natural products studied was found to increase with increasing concentration of the plant extracts. The main findings are summarized in Table 2

| S. no | Metals and their alloys | Green inhibitors. | Inhibitor concentration | Corrosive Medium | IE (%) | Methods | References |
|-------|-------------------------|-------------------|-------------------------|------------------|--------|---------|------------|
| 1     | Al                      | *Capparis decidua* (fruit, stem, and root extract) | 0.08-0.4 (unit not mentioned) | 0.5-5.0 N HCl, H₂SO₄ | 27.74-98.48 | Weight loss | 21 |
| 2.    | Al1060                  | *Jasmin nudiflorum* Lindl. (leaf extract) | 0.1-1.0 g/L | 1 M HCl | 69.0-93.6 | EIS | 22 |
| 3.    | Al5083                  | *Lawsonia inermis* (extract) | 200 ppm | Sea water | 45.50 | Weight loss, PDP, EIS | 23 |
| 4.    | Al                      | *Rosemary* (leaf extract) | 0.1-0.5 g/L | Biodiesel | 62.70-97.30 | Weight loss, PDP | 24 |
| 5.    | Al                      | Coffeehusk (extract) | 100-500 ppm | 0.5 M HCl | 48.90-92.70 | Weight loss, PDP, EIS, EFM, EIS | 25 |
| 6.    | Al1060                  | *Ananas communis* (leaf extract) | 0.1-0.5 g/L | 0.1 M HCl | 75.86-96.09 | Weight loss | 26 |
| 7.    | Al                      | *Ficus carica* (leaf extract) | 10-100 g/L | 0.5 M HCl | 58.65-91.34 | Weight loss | 27 |
| 8.    | Al1060                  | *Treculia Africana* (leaves extract) | 0.1-0.5 g/L | 1 M HCl | 41.30-94.00 | Weight loss, thermometric | 28 |
| 9.    | Al                      | *Dendrocalamus brandisii* (leaf) | 0.1-1.0 g/L | 1 M H₃PO₄ | 14.90-56.30 | PDP, EIS | 29 |
| 10.   | Al                      | *Citrus xanthocarpa* (leaves, seed, extract) | 0.12-0.60 | 0.5-5.0 N H₂SO₄ | 54.35-79.45 | Weight loss | 30 |
| 11.   | Al                      | *Ficus virens* (bark, leaf, seed) | 0.12-0.60 | 0.1-4.0 N HCl | 49.07-95.50 | Weight loss | 31 |
| 12.   | Al                      | *Ambrosia maritima* | 0.006-0.100 g/L | 0.5 M NaCl | 30.4-62.5 | PDP | 32 |
| 13.   | Al                      | *Carcinianodica* (seed extract) | 100-500 ppm | 0.5 M H₃PO₄ | 41.34-85.59 | PDP, EIS | 33 |
| 14.   | Al                      | *Neolamarckia cadamba* | 0.2-0.6 g/L | 1 M NaOH | 64.60-87.10 | Weight loss | 34 |
| 15.   | Al                      | *Senna auriculata* (leaf extract) | 300-900 ppm | 1 N NaCl | 52.20-76.20 | Weight loss | 35 |
| 16.   | Al                      | Saffron (leaf extract) | 600 ppm | 2 M HCL | 78.50-84.60 | Weight loss | 36 |
| 17.   | Al                      | *Solanum melongena* | 0.3-1.5 g/L | 1 M NaOH | 63.20-89.61 | Weight loss | 37 |
| 18.   | Al1060                  | Oliveseed (extract) | 1-10 vol% | 1 M HCl | 48.56-98.86 | Weight loss | 38 |
| 19.   | Al1060                  | *Vigna angularis* (husk extract) | 0.1-0.5 g/L | 0.5 M NaOH | 38.65-79.36 | Weight loss | 39 |
| 20.   | Al1060                  | *Trachyspermum ammicum* (seed extract) | 300-500 ppm | 0.5 M NaOH | 68.45-94.00 | Weight loss, PDP, EIS | 40 |
| 21.   | Al1060                  | *Gongronema latifolium* (leaf extract) | 10-50 vol % | 2 M HCl, KOH | 56.28-97.51 | HE | 41 |
| 22.   | Al                      | *Solanum melongena* | 0.6 g/L | 0.5 M | 16.25- | Weight loss | 42 |
Presently, the use of natural many plants extracts and various parts of neem plant extract as a green corrosion inhibitor has been summarized in Table 3 and 4. In this work, author is discussing about the many plant extract for inhibition of mild steel in acid test solution, and especially *Azadirachta indica* as green corrosion inhibitor for carbon steel, stainless steel, mild steel, Al, and zinc (Tables 3 and 4).

### Table 3: Inhibition of corrosion of mild steel by plants extract in different media.

| S. No | Plant                  | Corrosive solution | Methods                          | Temperature effect on inhibition | Concentration effect on inhibition | Reference |
|-------|------------------------|--------------------|----------------------------------|----------------------------------|-----------------------------------|-----------|
| 1.    | *E. crassipes* (leaf and root) | HCl                | Gasometric technique             | Decrease with rise in temperature | Increase                          | 47        |
| 2.    | *N. latifolia* (root)    | H₂SO₄              | Gasometric technique             | Decrease with rise in temperature | Increase                          | 48        |
| 3.    | Banana peel extract + Zn | Distilled water    | Weight loss, thermometric, FTIR | -                                | Decrease with Zn                  | 49        |
| 4.    | *Andrographispaniculata, Murraya koenigii* | HCl | DFT, molecule analysis | -                                | -                                 | 50        |
| 5.    | Aloe vera               | HNO₃               | Gravimetric technique            | Decrease with rise in temperature | Increase                          | 51        |
| 6.    | *C. sinensis* leaf extract | Aqueous           | Weight, pH, Uv-Vis, FTIR         | Decrease with rise in temperature | Decrease                          | 52        |
| 7.    | *H. rosa-sinensis* (Jasud) | HCl | Weight loss, Gravimetric, Electrochemical | Increase | Increase | 53        |
| 8.    | *I. paraguariensis*     | HCl                | Weight Electrochemical loss       | Increase                          | Increases                          | 54        |
| 9.    | *R. graveolens* extract | HCl                | Weight Electrochemical loss       | Decrease with rise in temperature | Increase                          | 55        |
| 10.   | *M. sapientum*          | H₂SO₄              | Weight Electrochemical loss       | Decrease                          | Increase                          | 56        |
| 11.   | *E. alba*               | H₂SO₄              | Weight Electrochemical loss       | Decrease with rise in temperature | Increase                          | 57        |
| 12.   | *T. catappa*            | HCl                | Weight Electrochemical loss       | Decrease                          | Increase                          | 58        |
| 13.   | *C. bracteosum*         | H₂SO₄              | Weight Electrochemical loss       | -                                | Increase                          | 59        |
| 14.   | *C. tetragonoloba*      | HCl                | Potentiodynamic Polarisation     | Decrease with rise in temperature | Increase                          | 60        |

Note: PDP-Potentiodynamic Polarisation; EIS-Electrochemical Impedance Spectroscop; HE-Hydrogen evolution
Inhibition of corrosion of metals by neem extracts:
Table 4 indicate that the inhibitory action of neem extract was reported by many investigators for inhibition of corrosion of aluminium, mild steel, carbon steel, stainless steel, coper and zinc in HCl, HNO₃ and H₂SO₄. Many investigators have been reported that the neem extract act as good green inhibitor for the protection of zinc, copper, Mild steel, carbon steel, stainless steel in acidic test solution. The inhibition of different metals corrosion by neem extract as a green inhibitor in nitric acid, hydrochloric acid and sulphuric acid solutions have been studied by many investigators using weight loss, gravimetric, kinetic activation and potentiodynamic polarisation techniques at various temperature. According to Ayssar et al., the neem extract was found to be an excellent potential green corrosion inhibitor for carbon steel in 1.0 M HCl. The inhibition efficiency increases with increasing the concentration of neem extract. Loto et al. study revealed that the corrosion inhibition efficiency of neem extract on the mild steel corrosion was increased in the dilute hydrochloric acid. The inhibition efficiency of the green inhibitor increased with increasing temperature, which clearly indicated the chemisorption of the green inhibitor on the surface of the metals. Almost in the all cases, neem extract act as good green corrosion inhibitor in the mineral acid test solution.

Green corrosion inhibitors mitigate the metal dissolution process by adsorption of natural ions or molecules onto metal/solution interface surface, minimizing the anodic and cathodic reaction at metal surface, by controlling the diffusion phenomenon for ionic species through the scale formed on the metal surface which also check the ionisation resistance on the active metal surface.

Table 4: Inhibition of corrosion of metals by neem extract (Azadirachtaindica) in acidic solution.

| S. No | Metals          | Corrosive solution | Methods                        | Temperature effect on inhibition | Concentration effect on inhibition | Reference |
|-------|-----------------|--------------------|--------------------------------|---------------------------------|------------------------------------|-----------|
| 1.    | Zn              | HCl                | Gravimetric and Thermometric   | Decrease                        | Increase                           | 61        |
| 2.    | Cu              | HNO₃               | Weight loss                    | Decrease                        | Increase                           | 62        |
| 3.    | Mild Steel      | HCl, H₂SO₄         | Gasometric Kinetic activation  | No Dependence                   | Increase                           | 63        |
| 4.    | Al              | HCl                | Potentiodynamic polarisation   | Decrease                        | Increase                           | 64        |
| 5.    | Mild steel      | H₂SO₄              | Weight loss                    | -                               | -                                  | 65        |
| 6.    | Mild steel      | HNO₃               | Gravimetric technique          | Decrease with rise temperature  | Increase                           | 66        |
| 7.    | Carbon steel    | HCl                | Weight loss                    | Decrease with rise temperature  | Increase                           | 67        |
| 8.    | Stainless steel | HCl, H₂SO₄        | Weight loss                    | Decrease at high temperature    | Increase                           | 68        |
| 9.    | Mild steel      | HCl, H₂SO₄        | Weight loss and Potentiodynamic| Decrease with rise temperature  | Increase                           | 69        |
| 10.   | Mild steel      | HCl                | Potentiodynamic polarisation   | Decrease with rise temperature  | Increase                           | 70        |

Conclusions:
Generally green corrosion inhibitors have excellent inhibiting efficiency under a variety of corrosive test solutions for metals. The non-toxicity and good biodegradability are the major advantages for these green inhibitors.

On the basis of above discussion, it is concluded that plant extracts are a good effective green corrosion inhibitor against several metals in various corrosive environment, especially for aluminium, mild steel, stainless steel, carbon steel and zinc. On the basis of published research work on the corrosion inhibition of metals and their alloys in different corrosive test solution, by several natural plant extract contain phytochemicals was presented in the present.
in this review. The various solvents like organic solvents, water, acids, and bases were used to extraction of extracts of various parts. The inhibition efficiency of the green corrosion inhibitors increased with increasing of concentration of constituents.

The inhibition efficiency of plant extract decreased with increasing the temperature. Weight loss was the classical method and frequently used to evaluate the corrosion inhibition efficiency of the studied plant extract as green inhibitors. Some studies were performed by electrochemical techniques. Physio-chemical adsorption of phytochemical was reported as the most probable mechanism on the surface of the metals. Alternatively, the inhibition action of these products was attributed to the formation of a protective barrier on the surface of the metals.

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