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STUDY ON CORROSION CHEMISTRY. PART-1: REVIEW ON THE ROLE OF VARIOUS EXTRACTS OF PLANT MATERIALS IN CORROSION

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ABSTRACT

The present review covers important aspects of the corrosion particularly green corrosion. It involves the seed, leaf etc. parts of the plant, in extract form, suitable for some inhibition to corrosion, were discussed to throw light on some aspects of corrosion. Its important is also discussed wherever available.

KEYWORDS: Seed, Leaf, Green corrosion and steel.

INTRODUCTION

During this period various books[1-3] were also published on the topic corrosion, but those are not that easily available to the researchers. Also, it is not possible for each author to cover all the details by new aspirants. It is a limitation of the book writing process, Hence we are writing the present review to add little updates to the readers of this journal. There is on-going collaboration between academia and industry, the book reviews[1] recent advances in research on oxide scale behavior in high-temperature forming processes. The book[3] on corrosion covers fundamental aspects of corrosion inhibition, historical developments and the industrial applications of inhibitors. Presenting novel, previously neglected approaches, the authors emphasize the pivotal role of reproducible experiments to elucidate the oxide scale properties and develop quantitative models with predictive accuracy. Each chapter consists of

a detailed, systematic examination of different aspects of oxide scale formation with immediate impact for researchers and developers in industry.

There is a review[4] and discussion on the theory and practical application of anti-corrosion coatings and their behavior. At the start two chapters reviewed the general requirements for inhibitors, classified them into types such as film-forming, adsorption, deactivation of medium (as oxygen or nitrous acid removal), passivation, extension of induction period; and discussed mechanism of inhibition. Further, more chapters dealt with inhibition in aqueous acids, aqueous bases, nearly neutral salt solutions, mixed or nonaqueous media, and protection against the atmosphere, particularly in "mothballing" or storage. Nearly 350 materials tested or used as inhibitors are noted in it, the more important ones being detailed and discussed in, with considerable data. Some commercial inhibitive preparations used in Soviet industry and nature are discussed, and also included the appendices given directions for using these in pickling and boiler cleaning particularly.

Rate of deposition and dissolution of iron has been measured as a function of potential for constant solution compositions.[5] Fe^{2+} concentration at constant pH; pH at constant Fe^{2+} concentration; and the presence of the anions SO_4^{2-}, Cl^-, ClO_4^-, Ac^- and NO_3^- at constant Fe_{2+} concentration. The measurement method was that of galvanostatic transients. Rate of hydrogen evolution reaction, both from H_3O^+ and H_2O, has been examined during these transients, as a function of potential and pH.

The cathodic slope (after correction for the partial current density due to H discharge) is apparently *RT/F* in (total) current density regions above those corresponding to the limiting current density for i_H. When account is taken of the pH change at the cathode surface due to H discharge, the cathodic Tafel slope becomes 2*RT/F*.

Several plants were tested (for different parts) for their performance in protecting steel and aluminum against corrosion under immersed conditions.[6] Calotropis procera, argemone mexicana, garlic and carrot were found to be good inhibitors for steel in neutral media, while black pepper, castor seed, tobacco, acacia gum and lignin proved to be good inhibitors for steel in acid media. For aluminum in acid medium, lignin, tobacco and black pepper were effective inhibitors.

The effects of various plant extracts on the dissolution of mild steel in HCl solutions were studied.[7] The additives investigated were *Papaia*, *Poinciana pulcherrima*, *Cassia occidentalis* and *Datura stramonium* seeds and *Papaia*, *Calotropis procera B*, *Azydracta indica* and *Auforpio turkiale* sap. Weight-loss determinations and electrochemical measurements were performed. It was found that all extracts except those of *Auforpio turkiale* and *Azydracta indica* reduced the corrosion of steel with an efficiency of 88%–96% in 1 N HCl and with a slightly lower efficiency in 2 N HCl. Both the cathodic evolution of hydrogen and the anodic dissolution of steel are inhibited. It is belief that the inhibition action is mostly due to the products of the hydrolysis of the protein content of these plants.

Various parts of several plants were tested for their performance in protecting steel and aluminum against corrosion under immersed conditions.[8] Argemone mexicana, calotropis procera, carrot and garlic were found to be good inhibitors for steel in neutral media, while tobacco, black pepper, castor seed, acacia gum and lignin proved to be good inhibitors for steel in acid media. For aluminum in acid medium, tobacco, lignin and black pepper were effective inhibitors.

The inhibitive effects of aqueous extracts of leaves of *Opuntia ficus indica* and *Aloe eru* and of the fruit-peels of Orange, Mango and Pomegranate[9] on the corrosion of mild steel, aluminum, zinc and copper in HCl and H_2SO_4 solutions have been investigated by means of weight loss and polarization measurements. The extracts retard the dissolution reactions to an extent dependent on the metal used, the concentration of the additive and the type, concentration and temperature of the attacking acid. The additives provide adequate protection to steel in 5 % HCl at 25 °C and in 10 % HCl at 25 °C and 40 °C. In the presence of a sufficient concentration of the extracts in 5 % HCl at 25 °C, the inhibitive efficiency towards steel decreases in the order: Mango (82%), Orange and Aloe eru (80 %), Opuntia (75 %), Pomegranate (65 %). The order of efficiency differs for the different metals, but extracting Mango peels is still the most effective for Al (82 %) and Zn (80 %). The most effective extract for Cu is that of Pomegranate fruit-shells (73 %). The extracts are generally more effective in HCl than in H_2SO_4.

The polarization measurements indicate that all the extracts increase the polarization of the cathodic reaction of all the four metals tested and the anodic polarization of steel and Zn, while Orange and Aloe have no effect on the anodic reaction of Al and only Pomegranate has

any effect on the anodic polarization of Cu. The results suggest that the extracts generally act as mixed inhibitors. The mechanism of inhibition has been discussed.

Factors establishing whether tannin and protein interact to form soluble complexes or precipitates were identified[10] and the ratio of tannin to protein in reaction mixture influenced solubility of complex (tannin-protein). When this ratio is larger than the optimum ratio, or equivalence point, soluble complexes(tannin-protein) are apparently formed instead of insoluble complexes. Other factors influenced the amount of protein precipitated by tannin-containing plant extracts, including length of reaction time and conditions of tannin extraction. Analytical and ecological significance of soluble complexes were considered. A titration method which allows simultaneous determination of the equivalence point and assessment of the protein-precipitating capacity of any plant extract was developed.

A general physicochemical definition of the term corrosion is given.[11] The main part of the document treats electrochemical corrosion of metals and alloys including uniform corrosion in the active and passive states and various types of nonuniform corrosion.

The system, carbon dioxide- water is of great scientific and technological importance. Thus, it has been studied often.[12] The literature for solubility of CO_2 in water is vast. An exhaustive survey was conducted and approximately 100 experimental investigations were found that reported equilibrium data at pressures below 1 MPa. A model based on Henry's law was used to correlate low pressure data. The correlation of Henry's constants is

$$\ln(H_{21}/\text{MPa}) = -6.8346 + 1.2817\times10^4/T - 3.7668\times10^6/T^2 + 2.997\times10^8/T^3$$

which is valid for $273<T<433$ K($0<t<160$ °C) where T is in K.

The review covers existing literature on solubility of carbon dioxide in water from 273 K to the critical temperature of solvent.[13] Results of the evaluation are expressed in the form of fitting equations for the infinite dilution Henry's constant, k^0, as a function of the density of the solvent, and also as an explicit function of the temperature. The pressure effect on the solubility is considered in the formulation. Different equations of state were used for the description of the CO_2-H_2O vapor phase and the effects on the calculated Henry's constant values are analyzed. The "best" solubility estimates are presented in smoothed tabular form.

The toxic corrosion inhibitor can be designed, if you have a good toxicity evaluation method of these molecules before these are actually synthesized.[14] A review of various procedures

of evaluation of aquatic toxicity of organic compounds and highlights the relationship between the structures of these inhibitors and their aquatic toxicity. Such relationships can form the basis for changing the structure of the existing corrosion inhibitors to make these less toxic.

Electrochemical noise analysis was used to continuously monitor the film formation and destruction processes of CO_2 corrosion inhibitor imidazoline.[15] It is an inhibitor commonly used for protecting wells(gas and oil) and flow-lines from carbon dioxide corrosion. The experimental results show that trends in electro-chemical noise effectively followed inhibitor film formation and destruction processes. The electrochemical noise data analysis strongly suggests that Electrochemical noise analysis is a practical technique in the continuous monitoring of inhibitor film performance and in the evaluation of inhibitor film. The electrochemical noise resistance is confirmed to be strongly correlated to polarization resistance, although the theoretical background and data analysis methods. Electrochemical noise analysis is also shown to be a convenient method for continuous corrosion rate monitoring.

Industrial processes for extraction of oil from edible oilseeds generally involve solvent extraction steps which may or may not be preceded by pressing.[16] Useful and preferred solvent is hexane and the process had been in commercial operation for a long time. It is possible to achieve oil yields in excess of 95% with a solvent recovery of over 95%. In the past, the main concern of this process has been the safety implications surrounding the use of hexane. To develop processes based on the use of aqueous extraction media which were unsuccessful mainly due to low oil yields. Interest in aqueous extraction processes has been revived by increasing environmental concern. An aqueous process is looked upon as an environmentally cleaner alternative technology for oil extraction. Organic solvents such as hexane, in particular, can contribute to the industrial emissions of volatile organic compounds. The production of volatile organic compounds in conventional processes is particularly worrisome since these can react in the atmosphere with other pollutants to produce ozone and other photochemical oxidants which can be hazardous to human health and can cause damage to crops. Besides this, the volatile organic compounds are themselves “greenhouse gasses” some are carcinogenic and have toxic properties. Other advantages of the aqueous process compared with solvent-based processes include.

(1) simultaneous production of edible oil and protein isolate or concentration in the same process.
(2) lower protein damage during extraction, and
(3) improved process safety due to the lower risk of fire and explosion. It is also reported that aqueous extraction processes may be more cost effective since the solvent recovery step is eliminated. The main limitations of this process appear to be:
(1) lower efficiency of oil extraction as evident in earlier studies,
(2) demulsification requirements to recover oil when emulsions are formed, and
(3) treatment of the resulting aqueous effluent.

With the objective of improving the yield of aqueous processes, enzymes have been used to facilitate oil release. Selected enzymes have been tried on different types of oilseeds, resulting in extraction yields much higher than the original aqueous process. These enzymes mainly hydrolyze the structural polysaccharides which form cell walls of oilseeds or proteins which form cell and lipid body membranes. This article aims to review aqueous and enzyme-based processes and discuss related issues.

Many different models for CO_2 corrosion are used by engineers in the oil and gas industry.[17] Some are described in open literature and only those are covered by this review. In order to classify carbon dioxide corrosion models, we have arbitrarily decided to group them into three categories based on how firmly they are grounded in theory: mechanistic, semi-empirical and empirical models. A few most representative examples are discussed from each group. Performance of four different models covering all three groups is tested by comparing predictions with a large experimental carbon dioxide corrosion database.

Efficiency of corrosion inhibitor by acid extract of dry Emblica officinalis leaves for mild steel in 1N HCl medium is investigated.[18] Wherein the experimental methods include potentiodynamic polarization, weight loss and impedance studies. Results showed Emblica officinalis leaves to be a good corrosion inhibitor of a mixed type and having efficiency of 87 % at 2 % v/v concentration of inhibitor. Corrosion inhibition may be due to the spontaneous physical adsorption of the plant constituents on the mild steel surface. Experimental data also fitted for Freundlich, Langmuir, Temkin and Flory-Huggins adsorption isotherms.

The inhibitive effect of damsissa (Ambrosia maritime, L.) plant extracts on the corrosion of steel in an aqueous solution of 1 M sulfuric acid (H_2SO_4) was investigated[19] in the

temperature range from 25°C to 40°C using potentiodynamic polarization and electrochemical impedance spectroscopy techniques. The stability of the inhibition efficiency of damsissa extracts with immersion time was examined by weight-loss measurements at 30°C and 60°C. Potentiodynamic polarization curves indicated that the damsissa extract behaves as a mixed-type inhibitor. EIS measurements showed that the dissolution process occurs under activation control. Corrosion rates of steel and efficiencies of inhibition of extract obtained from polarization and impedance measurements are in good agreement. Inhibition was found directly proportional to the concentration of the plant extract but decreases with increasing temperature. The results obtained show that the damsissa extract could serve as an effective inhibitor for corrosion of steel in H_2SO_4 media. Inhibitory actions of damsissa extract are criticized as per the adsorption of molecules (on electrode surface through the active centers). Theoretical fitting of different isotherms, Frumkin, Langmuir, Flory-Huggins and the kinetic-thermodynamic model, were tested to clarify nature of adsorption. Activation parameters associated were determined and discussed.

Corrosion rate data for several commercial carbon dioxide corrosion inhibitors[20] have been fitted using van't Hoff equation, and Temkin adsorption isotherm enabling a determination of the enthalpy of adsorption (ΔH_{ad}°). It has been found that experimentally determined ΔH_{ad}° values for commercial inhibitor formulations can provide valuable insights into behavior of inherent active components. Sensitivities of temperature along with the mechanism of physi- or chemi-sorption and the minimum concentration required for inhibitors activity is determinable by this method. The FTIR has been used to identify the inhibitors which are adsorbed tenaciously on the steel and suitable for use in batch wise treatment.

Cashew Nut Shell Liquid has been tested[21] as a corrosion inhibitor for carbon steel in 3 % aqueous NaCl solution (pH 6) saturated with carbon dioxide gas at 30 °C under static conditions using ac-impedance and potentiodynamic polarization techniques. It was found that Cashew Nut Shell Liquid reduces the extent of the electrochemical processes taking place on carbon steel undergoing corrosion. The corrosion rate of the carbon steel was reduced by over 92 % when only 300 ppm of Cashew Nut Shell Liquid was applied. This indicates that Cashew Nut Shell Liquid is a potential corrosion inhibitor for carbon steels in the CO_2 environment.

The inhibitive action of the mucilage extracted from the modified stems of prickly pears, toward acid corrosion of aluminum, is tested using weight loss, thermometry, hydrogen evolution and polarization techniques.[22] It was found that the extract acts as a good corrosion inhibitor for aluminum corrosion in 2.0 M HCl solution. The inhibition action of the extract was discussed in view of Langmuir adsorption isotherm. It was found that the adsorption of the extract on aluminum surface is a spontaneous process. The inhibition efficiency increases as the extract concentration is increased. The effect of temperature on the IE was studied. It was found that the presence of extract increases the activation energy of the corrosion reaction. Moreover, the thermodynamic parameters of the adsorption process were calculated. It was found also that the *Opuntia* extract provides a good protection to aluminum against pitting corrosion in chloride ion containing solutions.

A specially designed electrochemical cell incorporating a rotating disc electrode has been used for *in situ* surface-enhanced Raman spectroscopy studies of the adsorption of inhibitors of carbon dioxide corrosion onto silver-coated mild steel electrodes.[23] It is shown that surface-enhanced Raman spectroscopy-active inhibitors comprising aromatic moieties may be detected using the surface-enhanced Raman spectroscopy technique. Furthermore, the efficacy of adsorption of corrosion inhibitors employed in the present study is optimal near the open cell or corrosion potential, demonstrating that electrode polarization induces electrostatic forces of repulsion that retard the adsorption of the inhibitor to the corroding steel surface.

Extraction of anthocyanins from black currants using aqueous ethanol was optimized for yield and antioxidant activity.[24] The process variable having the most effect on the extraction was the solvent to solid ratio, which increased phenolic extraction in the whole range from 0 to 19 L/kg. Total phenolics increased with ethanol concentration up to a maximum at about 60 % and then decreased with further increase in solvent concentration irrespective of the solvent to solid ratio. Temperature only affected the extraction of anthocyanins. Increasing the temperature beyond 30 to 35 °C resulted in degradation of anthocyanins and reduction of yields. Variation in extract composition was not sufficiently large to affect antioxidant activity.

Influence of addition of the naturally occurring substance henna on the corrosion of iron in molar hydrochloric acid was studied by weight loss measurement.[25] The Henna substance reduces corrosion rate. Inhibition efficiency directly proportional to henna concentration.

Effect of temperature on corrosion behavior of iron indicated that inhibition efficiency decreased with temperature. Adsorption of henna on iron was found to follow the Langmuir isotherm of adsorption.

A semiautomatic method based on application of ultrasounds has been developed to leach and hydrolyse phenolic compounds, such as rutin, naringin, ellagic acid, naringenin, kaempferol and quercetin from strawberries.[26] Two grams of lyophilized sample was placed into a sample cell and 5 mL of acetone containing HCl was added. The cell was immersed in a water bath and sonicated for 30 s for 3 times: each time 5 mL extractant displaced the previous extract. On the completion of extraction, combined extracts were evaporated for 10 min, diluted to 10 mL with water adjusted to pH 8, and transferred to a cleanup-preconcentration manifold; here the analytes were retained in two in-series mini-columns packed with HR-P sorbent and then eluted with 4 mL methanol, and injected for individual separation-quantitation into a chromatograph-photodiode array detector(PDA) assembly. Optimisation of the extraction was carried out using samples spiked with 4 mg kg^{-1} of each analyte. The calibration curves using standard addition in red strawberries typically gave linear dynamic ranges of 4-40 mg L^{-1} for all analytes, except for ellagic acid (40-400 mg L^{-1}). The r(2) values exceeded 0.98 in all studied samples.

Inhibition of corrosion of aluminum in hydrochloric acid in the presence of Carica papaya and Azadirachta indica at 30-40°C was studied using the weight loss, thermometric and hydrogen evolution techniques.[27] The inhibition efficiency (% Inhibition) increased with increase in concentration of the extracts and with increase in temperature. Carica papaya is a better inhibitor at 30°C whereas Azadirachta indica is better at 40°C. Carica papaya and Azadirachta indica were found to obey Temkin, Freundlich and Flory-Huggins adsorption isotherms at all concentrations studied at 40°C. The phenomenon of chemical adsorption is proposed from the obtained Ea, ΔG_{ads} and Q_{ads} values calculated.

Inhibition effect of Zanthoxylum alatum plant extract on corrosion of mild steel in 20, 50 and 88% aqueous orthophosphoric acid has been investigated[28] by weight loss and electrochemical impedance spectroscopy. The plant extract is able to reduce the corrosion of steel more effectively in 88% phosphoric acid than in 20 % phosphoric acid. The effect of temperature on the corrosion behavior of mild steel in 20, 50 and 88 % phosphoric acid with addition of plant extract was studied in the temperature range 50–80 °C. Results on corrosion rate and inhibition efficiency have indicated that this extract is effective up to 70 °C

in 88 % phosphoric acid medium. Surface analysis viz. FT-IR and XPS was also carried out to establish the mechanism of corrosion inhibition of mild steel in phosphoric acid medium.

An aqueous extract of plant material rhizome of Curcuma longa L. powder has been employed as a corrosion inhibitor in controlling corrosion[29] of carbon steel immersed in the aqueous solution containing 60 ppm of Cl^-, by the weight-loss method, in absence and presence of Zn^{2+}. Important constituent of this plant extract is curcumin. It has good inhibition efficiency and shows it is good at extreme pH values while in the presence of Zn^{2+}, there exists a synergistic effect. Synergism parameters have been calculated. The protective film has been analyzed using Fourier transform infrared spectroscopy. The film consists of a Fe^{2+}-curcumin complex and zinc hydroxide ($Zn[OH]_2$). It is found to be UV-fluorescent. Electrochemical studies such as potentiodynamic polarization and alternating current impedance have been used to study the mechanism characteristics of corrosion inhibition.

The grape byproducts which are a cheap source of dietary supplements extractable as food preservatives or antiradical phenols, for disease prevention.[30] The efficiency of extraction strongly depends on operative conditions. The effects of three critical variables –sample quantity, flow-rate and particle size – on extraction efficiency were also studied. The process conditions maximize the activity (antiradical) of phenolic extract. The extraction promoted formation of phenolic compounds not naturally occurring in grape residue. Chromatographic profiles of extracts confirmed that these novel compounds were formed as a consequence of phenolic polymer formation.

Aqueous extract of leaves of henna (lawsonia) is tested as corrosion inhibitor of C-steel[31], nickel and zinc in acidic, alkaline and neutral solutions, using polarization technique. It was noted that the extract acts as a good corrosion inhibitor for 3 tested electrodes in all media tested. Inhibition efficiency is directly proportional. The degree of inhibition depends on the nature of metal and the type of medium. For C-steel and nickel, inhibition efficiency increases in the order: alkaline < neutral < acid, while for zinc it increases in the order: acid < alkaline < neutral.

Extract acts as a mixed inhibitor. Inhibitive action of the extract is discussed in view of adsorption of lawsonia molecules on metal surface. It was found that this adsorption follows Langmuir adsorption in all systems tested. Formation of complexes between M(metal)

cations and lawsone is also discussed as an additional inhibition mechanism of C-steel and nickel corrosion.

The extracts of plant materials are environmentally friendly and less expensive and contain many active principles.[32] The polar atoms such as S, N, O, P etc. Due to this nature, the lone pair of electrons present on these atoms is pumped onto the metal surface; loss of electrons from the metal surface can be avoided. Hence corrosion inhibition takes place. Due to adsorption of inhibitor molecules on metal surfaces, protective film is formed and is thus controlled by corrosion.

The inhibitory effect of extracts of Allium sativum in the corrosion of mild steel in H_2SO_4 solutions was studied using weight loss method[33] and hydrogen evolution at room temperature. The results reveal that plant extract acts as an inhibitor for the corrosion of mild steel in H_2SO_4 solutions. Inhibition efficiency is directly proportional to extract concentration. Langmuir isotherm as well as the thermodynamic-kinetic model of El-Awady et al. was tested to their fit to the experimentally obtained data. The free energies and equilibrium constant for the adsorption process were determined. A possible mechanism of adsorption is discussed.

In chloride solutions, tinplate exhibits localized corrosion processes due to the defects or imperfections of the outer Sn layer, which leaves the Sn–Fe alloyed layer exposed to aggressive solutions.[34] The anodic character of the external Sn with respect to the internal alloyed layer, will lead to its dissolution acting as a sacrificial anode. The addition of cerium salts to the aggressive media decreases the pitting susceptibility of tinplate by means of cerium precipitation on the cathodic sites (bare Sn–Fe areas). This induces a change of the controlling mechanism suggesting that cerium is a cathodic inhibitor for tinplate, which could be corroborated by removal selectively the outer Sn layer and leaving the Sn–Fe layer directly exposed to the solution. In these conditions, the corrosion process of the steel base will take place through the defects of the Sn–Fe layer, which presents a cathodic character in comparison to the base metal, inducing the precipitation of cerium on the alloyed layer.

The extract from the seeds of Garcinia kola was investigated as a corrosion inhibitor for mild steel in 2M HCl and 1M H_2SO_4 solutions using the gasometric technique.[35] The results indicate that the extract inhibited the metal corrosion in the acidic environments and inhibition efficiency increased with concentration. Temperature studies revealed a decrease in

efficiency with rise in temperature and corrosion activation energies increased in the presence of the extract. A mechanism of physical adsorption is proposed for the inhibition behavior.

The inhibitive action of seed extracts of *Garcinia kola* on mild steel corrosion in 2M HCl and 1M H_2SO_4 solutions was studied using the gasometric technique.[36] The results indicate that the extract functioned as a good inhibitor in the acidic environments and inhibition efficiency increased with concentration. Temperature studies revealed a decrease in efficiency with rise in temperature and corrosion activation energies increased in the presence of the extract. A mechanism of physical adsorption is proposed for the inhibition behaviour. Potassium iodide additives had a beneficial effect on the efficiency of the extract.

The corrosion inhibition of aluminum in sulfuric acid solution in the presence of different plant parts, namely, leaves, latex, and fruit was studied[37] using weight loss method and thermometric method. The ethanolic extracts of *Calotropis procera* and *Calotropis gigantea* act as an inhibitor in the acid environment. The inhibition efficiency increases with increase in inhibitor concentration. The plant parts inhibit aluminum, and inhibition is attributed, due to the adsorption of the plant part on the surface of aluminum.

The alcoholic extracts of stem bark, leaves and fruit from the Prosopis cineraria are tested for their effectiveness to combat corrosion of mild steel in hydrochloric acid, sulphuric acid and in acid mixture (hydrochloric acid sulphuric acid) by the mass loss method.[38] It was found that the presence of the extracts reduces the corrosion rate of mild steel in acidic solution. The inhibition efficiency increases as the extract concentration is increased. The fruit extract of Prosopis cineraria shows a maximum corrosion inhibition efficiency in the acid mixture solution compared to leaves and stem bark extracts. The Prosopis extract provides a good protection to mild steel against corrosion.

In this research the attempts were made to utilize the aqueous extract of natural compound, namely Date Pits (DP, hard stone of date palm) as acid corrosion inhibitor[39] for mild steel. The inhibition efficiency is evaluated by different techniques; as weight loss, polarization measurements (Tafel plot and Linear polarization) and scanning electron microscope.

The effect of extracts of Chamomile (*Chamaemelum mixtum* L.), Halfabar (*Cymbopogon proximus*), Black cumin (*Nigella sativa* L.), and Kidney bean (*Phaseolus vulgaris* L.) plants[40] on the corrosion of steel in aqueous 1 M sulphuric acid were investigated by

electrochemical impedance spectroscopy and potentiodynamic polarization techniques. EIS measurements showed that the dissolution process of steel occurs under activation control. Potentiodynamic polarization curves indicated that the plant extracts behave as mixed-type inhibitors. The corrosion rates of steel and the inhibition efficiencies of the extracts were calculated. The results obtained show that the extract solution of the plant could serve as an effective inhibitor for the corrosion of steel in sulphuric acid media. Inhibition was found to increase with increasing concentration of the plant extract up to a critical concentration. The inhibitive actions of plant extracts are discussed on the basis of adsorption of stable complexes at the steel surface. Theoretical fitting of different isotherms, Langmuir, Flory–Huggins, and the kinetic–thermodynamic model, were tested to clarify the nature of adsorption.

To find corrosion inhibitors for acid corrosion processes that not only are highly effective[41] but also more acceptable to the environment than conventional types of inhibitor compounds, a performance screening of several natural products was carried out for carbon steel corrosion inhibition in 5% HCl acid solution at different temperatures using weight loss and electrochemical methods (Tafel line and Polarization resistance). Also, the morphology of the surfaces of samples tested was examined by scanning electron microscope. All the investigated natural compounds exhibited excellent corrosion inhibition properties for carbon steel in 5% HCl solution.

The inhibitive effect of the extract of khillah (*Ammi visnaga*) seeds, on the corrosion of SX 316 steel[42] in HCl solution was determined using weight loss measurements as well as potentiostatic technique. It was found that the presence of the extract reduces markedly the corrosion rate of steel in the acid solution. The inhibition efficiency increases as the extract concentration is increased. The inhibitive effect of khillah extract was discussed on the basis of adsorption of its components on the metal surface. Negative values were calculated for the energy of adsorption indicating the spontaneity of the adsorption process. The formation of insoluble complexes as a result of interaction between iron cations and khellin, which present in the extract, was also discussed.

Natural oil extracted from Mentha pulegium, was evaluated as a corrosion inhibitor of steel[43] in molar hydrochloric using weight loss measurements, electrochemical polarization and EIS methods. The naturally oil was found to retard the corrosion rate of steel. The inhibition efficiency was found to increase with oil content to attain 80 % at 2.76 g/L. Mentha

pulegium oil acts as a cathodic inhibitor. The increase in temperature leads to an increase in the inhibition efficiency of the natural substance. The adsorption isotherm of natural products on the steel has been determined.

The first report on the cost of corrosion in Japan was published in 1977.[44] The report estimated that the corrosion loss in Japan which did not include indirect loss was 1-2 percent of Gross National Product at that time. Since then, almost two decades have passed and the industrial structure has drastically changed. Corresponding to this situation, the Committee on the Cost of Corrosion in Japan was organized in 1999 jointly by the Japan Society of Corrosion Engineering and the Japan Association of Corrosion Control (JACC). The project was funded by the National Research Institute for Metals as part of the STX-21 Project. The cost of corrosion in 1997 was estimated by the Uhlig method and the Hoar method. The estimated cost was compared with the past data which was estimated in 1974 by the same methods of the Uhlig and the Hoar method. In addition to the above estimation, a preliminary analysis by the Input/Output method was performed to estimate the total cost of corrosion including the direct and indirect costs. The overall costs estimated by the Uhlig and Hoar methods for 1997 were 3,938 billion yen and 5,258 billion yen, respectively, which were equivalent to 0.77 % and 1.02 % of the GNP of Japan. The total cost including the direct and indirect costs, which were estimated preliminary by the Input/ Output analysis, is likely to be more than 2 times larger than the direct cost estimated by the Uhlig method.

The inhibitive behavior on steel of flavonoid monomers[45] that constitute mangrove tannins namely catechin, epicatechin, epigallocatechin and epicatechin gallate was investigated in an aerated HCl solution via electrochemical methods. The monomers were found to be mainly cathodic inhibitors and the inhibition efficiency was dependent on concentration. To explain the adsorptive behavior of the molecules on the steel surface, a semiempirical approach involving quantum chemical calculations using HyperChem 6.0 was undertaken. The HOMO electronic density of the molecule was used to explain the inhibiting mechanism. The most probable adsorption centers were found in the vicinity of the phenolic groups. In a second part, the use of mangrove tannin, extracted from the mangrove barks as steel corrosion inhibitors in acidic media was investigated and its inhibitive efficiency was compared with that of commercial mimosa, quebracho and chestnut tannins. The inhibitive performance of mangrove tannins was comparable to the other tannins investigated, indicating their potential in corrosion protection.

The state-of-the-art in modeling of internal corrosion of oil and gas pipelines made from carbon steel is reviewed.[46] The review covers the effects of: electrochemistry, water chemistry, formation of protective scales and scales, temperature, flow, steel, inhibition, water condensation, glycol/methanol and localized attack. Various mathematical modeling strategies are discussed.

Azadirachta indica leaves[47] extract was investigated as a copper corrosion inhibitor in 0.5 M sulphuric acid. Inhibition efficiency of *Azadirachta indica* was compared to that of the already proven good inhibitors 2-acetamino-5-mercapto-1,3,4-thiadiazole and 1,2,3-benzotriazole. The inhibition properties were studied using electrochemical polarization and weight loss techniques. In the region of active copper dissolution, the highest inhibition efficiency was exhibited by (92.7 %). *Azadirachta indica* exhibited somewhat higher efficiency (86.4 %) than the widely used (85.5 %), showing that the extract could serve as an effective substitute for currently preferred copper corrosion inhibitors in sulphuric acid. The weight loss results were interpreted by means of the Frumkin isotherm of adsorption on the metal surface. The values of ΔGads equal to − 41.96 kJ mol^{-1} for 2-acetamino-5-mercapto-1,3,4-thiadiazole and 1,2,3-benzotriazole. and − 35.22 kJ mol^{-1} for compound under study indicate strong spontaneous adsorption while the surface coverage dependence on the log *c* following the Frumkin isotherm is suggestive of chemisorption in case of all three tested inhibitors.

Solvents define a major part of the environmental performance of processes in the chemical industry and also impact on cost, safety and health issues.[48] The idea of “green” solvents expresses the goal to minimize the environmental impact resulting from the use of solvents in chemical production. Here the question is raised of how to measure how “green” a solvent is. The framework combines the assessment of substance-specific hazards with the quantification of emissions and resource use over the full life-cycle of a solvent. A comprehensive framework for the environmental assessment of solvents that covers major aspects of the environmental performance of solvents in chemical production, as well as important health and safety issues is proposed in this article. The proposed framework is demonstrated on 26 organic solvents. Results show that simple alcohols (methanol, ethanol) or alkanes (heptane, hexane) are environmentally preferable solvents, whereas the use of dioxane, acetonitrile, acids, formaldehyde, and tetrahydrofuran is not recommendable from an environmental perspective. The present application demonstrates that the presented

framework is a useful instrument to select green solvents or environmentally sound solvent mixtures for processes in the chemical industry.

Inhibitive action of aqueous extract of olive (*Olea europaea L.*) leaves in case of corrosion of C-steel in 2 M HCl solution was studied applying the loss in weight measurements, cyclic voltammetry and Tafel polarization.[49] It was marked that extract is useful as a good corrosion inhibitor for the tested system. Inhibitive efficiency is directly proportional to the leaf extract concentration. Inhibitive action of extract is discussed with a view to adsorption of its components onto the surface of the steel, making a barrier to mass and charge transfer. It was found also that such adsorption increases the activation energy of the corrosion process. The adsorption of extract components onto the steel surface was found to be a spontaneous process and observed to follow the Langmuir adsorption isotherm. Results of cyclic voltammetry indicated that presence of olive extract is inversely proportional to charge density in transpassive regions. Inhibition efficiency is inversely proportional to as the temperature is increased.

To evaluate the effect of different parts of *Carica papaya* (leaves, seeds, heartwood and bark) as eco- friendly and non- toxic mild- steel corrosion inhibitors in H_2SO_4 at 30 to 60 ^{0}C.[50] In this article the acid extracts of the different parts of *Carica papaya* were used as inhibitors in various corrosion tests. Gravimetric and gasometric techniques were used to characterize the mechanism of inhibition. The leaves, seeds, heartwood and bark extracts were found to inhibit mild steel corrosion in H_2SO_4. The inhibition efficiencies of the plant's part extracts follow the trend.

leaves > seeds > heartwood > bark

Inhibition efficiency is directly proportional to the concentration of extracts but inversely proportional to the temperature. Physical adsorption of the phytochemical components of the plant on the metal surface is proposed as the mechanism of inhibition. The experimental data fits well into the Temkin and Langmuir adsorption isotherms. The plant extracts can be used in chemical cleaning and picking processes. This research provides information on the possible use of the different parts of *Carica papaya* as sources of cheap eco- friendly and non- toxic corrosion inhibitors.

The inhibitive action of leaves, seeds and a combination of leaves and seeds extracts of *Phyllanthus amarus* on mild steel corrosion[51] in HCl and H^2SO^4 solutions was studied using

weight loss and gasometric techniques. Results indicate that the extracts functioned as a good inhibitor in both environments and inhibition efficiency increased with extracts concentration. Temperature studies revealed an increase in inhibition efficiency with rise in temperature and activation energies decreased in the presence of the extract. A mechanism of chemical adsorption of the plant's components on the surface of the metal is proposed for the inhibition behavior. The adsorption characteristics of the inhibitor were approximated by Temkin isotherm.

The work aims to investigate the inhibitive effect of ethanol extracts of *Garcinia kola*[52] for the corrosion of mild steel in H2SO4 solutions. The study is another trial to find a cheap and environmentally safe inhibitor for mild steel corrosion. The inhibition efficiency has been evaluated using the hydrogen evolution technique at 30- 60°C. The mechanism of adsorption inhibition and type of adsorption isotherm were proposed based on the trend of inhibition efficiency and kinetic data. The results obtained indicate that extracts of *Garcinia kola* inhibits the corrosion of mild steel in acidic medium and that the inhibition efficiency increases with an increase in the concentration of ethanol extracts and decreasing temperature. The inhibition efficiency increased in addition of potassium iodide to extracts of *Garcinia kola*, indicating synergism. The experimental data obeyed the Langmuir adsorption isotherm and it is a thermodynamic- kinetic model. The activation energy of inhibition of 6.8508 KJ/mol calculated for the corrosion process suggests that the extracts of *Garcinia kola* molecules are physically adsorbed on the metal surface. Further investigations involving electrochemical studies such as polarization method will provide further enlightenment on the mechanistic aspect of corrosion inhibition. This work provides new information on the possible application of extracts of *Garcinia kola* as an environmentally friendly corrosion inhibitor under the specified conditions. This environmentally friendly inhibitor could find possible applications in metal surface anodizing and surface coatings.

The inhibition of the corrosion of mild steel by ethanol extract of *Musa sapientum* peels in H_2SO_4 has been studied using gasometric and thermometric methods.[53] The results of the study reveals that the different concentrations of ethanol extract of *M. sapientum* peels inhibit mild steel corrosion. Inhibition efficiency of the extract is found to vary with concentration, temperature, period of immersion and pH. Values of activation energy of the inhibited corrosion reaction of mild steel are greater than the value obtained for the blank. Thermodynamic consideration reveals that adsorption of *M. sapientum* peels extract on mild

steel surface is spontaneous and occurred according to Langmuir and Frumkin adsorption isotherms. A physical adsorption mechanism has also been proposed for the adsorption of the inhibitor.

The use of inhibitors for the control of corrosion of metals and alloys which are in contact with an aggressive environment is an accepted practice. Large numbers of organic compounds were studied and are being studied to investigate their corrosion inhibition potential.[54] All these studies reveal that organic compounds especially those with N, S and O showed significant inhibition efficiency. But, unfortunately most of these compounds are not only expensive but also toxic to living beings. It is needless to point out the importance of cheap, safe inhibitors of corrosion. Plant extracts have become important as an environmentally acceptable, readily available and renewable source for a wide range of inhibitors. They are rich sources of ingredients which have very high inhibition efficiency. This article gives a vivid account of natural products which are used as corrosion inhibitors for various metals and alloys in aggressive media.

The inhibitive action of Tagetes erecta-leaves[55] extract on mild steel in 1 M HCl has been studied using weight loss, electrochemical polarization and AC impedance measurements and The extract shows a very good inhibition in the hydrochloric acid medium and inhibition efficiency of the extract was found to vary with the extract concentration and immersion time. The maximum inhibition efficiency of 96 % was obtained at 0.3 % v/v concentration of the extract at 24h immersion time at room temperature. The potentiodynamic polarization data showed that the extract is of mixed type. EIS measurements showed that the dissolution process occurs under activation control. The protective film formed on the surface of mild steel by the adsorption of phytochemical constituents present in the extract was confirmed by SEM studies.

The inhibitory effect of Bauhinia purpurea (Fabaceae) extract was studied in the corrosion of carbon steel in 1.0 $mol \cdot L^{-1}$ sulfuric acid solution.[56] This plant was collected from the Brazilian rainforest. The carbon steel protection was observed by varying the extract concentration from 50 to 500 $mg \cdot L^{-1}$. Polarization curves revealed that this extract acted as an adsorption inhibitor decreasing both anodic and cathodic density currents. Weight loss measurements showed that the extract remains stabler at least 72 hours. The adsorption process of this extract obeyed the Langmuir adsorption isotherm. Lastly, Arrhenius' plot suggested the physical adsorption of the extract.

Corrosion inhibition of mild steel in 1 M HCl was investigated in the absence and presence of different concentrations of extract of Crossandra infundibuliformis leaves.[57] Weight loss measurements and electrochemical studies were employed. The corrosion rate of mild steel and the inhibition efficiencies of the extract were calculated. The results obtained show that the extract solution of the plant could serve as an effective inhibitor for the corrosion of mild steel in HCl media. Inhibition was found to increase with increasing concentration of the plant extract. The inhibitive actions of plant extract are discussed on the basis of adsorption of stable complexes at the mild steel surface. Theoretical fitting of different isotherms, Langmuir, Temkin, Freundlich, Frumkin, Flory-Huggins and the kinetic thermodynamic model were tested to clarify the nature of adsorption. Polarization curves revealed that this inhibitor acts as a mixed type inhibitor and the inhibition efficiency 97 % can be obtained. The surface analysis study confirms the corrosion of mild steel and its inhibition by the inhibitor.

Rosmarinus officinalis essential oil obtained by hydrodistillation was tested as a corrosion inhibitor of steel in 0.5M H_2SO_4 using weight loss measurements, and electrochemical polarization methods.[58] Results obtained indicate that the corrosion rate is reduced and R. officinalis oil adsorbs on the metal surface and then inhibits corrosion process. Its inhibition efficiency increases with oil increasing content to reach 61% at 1 g/L. Polarisation curves indicate that RO acts as a cathodic inhibitor. The inhibition efficiency of naturally oil remains slightly constant with the rise of temperature. Chemical analysis of R. off icinalis essential oil was carried out using capillary GC and GC/MS. The oil composition was characterized by a high amount of oxygenated monoterpene (77 %) with 1,8-cineole (52.1 %) as the main component. Discussion of adsorption has been investigated on the basis of essential oil composition. Finally, the isotherm adsorption of essential oil on the steel has been also reported.

CONCLUSION

The present review updates the progress of green corrosion chemistry for the steel and few papers on aluminium materials. It will be useful to green corrosion chemist and researchers in the near future.

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