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Green Corrosion Inhibitors for Zinc -An Overview

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ABSTRACT: Corrosion can be defined as a phenomenon of physicochemical interaction of a metal and its environment, inducing degradation of the metal itself. Corrosion can be controlled or minimized by the use of inhibitors. To remedy this problem, laboratory research has shown the great interest in using essential oils and extracts of aromatic and medicinal plants, which are natural, non-toxic and renewable products, as corrosion inhibitors. In this review paper, corrosion inhibition of zinc by green inhibitors in various media have been reported. The inhibition's efficiency of inhibitor compounds is strongly dependent on the structure and the chemical properties of the film formed on the metal surface. The protection of zinc metal from corrosion is analyzed by many technologies such as weight loss (Gravimetric), temperature effect, Gasometric, Potentiodynamic polarization and Electrochemical Impedance Spectroscopy (EIS). The protective films formed on metal surface have been analyzed by various techniques such as Scanning Electron Microscopy (SEM), Atomic Force Microscopy (AFM), FT-IR (Fourier Transform-Infrared Spectroscopy),Ultra-Violet(UV)Visible Spectra, X-ray Diffraction Spectroscopy (XRD), Energy Dispersive X-ray Spectroscopy (EDX), Infra-red (IR), Electrochemical Frequency Modulation (EFM), Cyclic Voltametry and adsorption study.

KEYWORDS: Corrosion, Zinc, Green inhibitors, Potentiodynamic Polarization, EIS, SEM.

I. INTRODUCTION

Corrosion is defined as destruction or deterioration of a material (metal) because of its reaction with environment. Zinc is an important metal with numerous industrial applications [1-3] and is mainly used for the corrosion sacrificial protection of steel [4-7]. Zn and its alloys are used in large scale, its alloys are corrosive in nature and it easily corrodes under the mild conditions. Due to the increasing usage of zinc, the study of corrosion inhibition is most important one. One of the methods used to reduce the rate of metal corrosion is the addition of inhibitors. A corrosion inhibitor is "a chemical substance which, when added at low concentration to the corrosive medium, slows or stops the corrosion process of a metal in contact with that medium [8].

Several studies have indicated that most organic and inorganic inhibitors are toxic and pose serious hazards to humans and the environment during their applications [9,10]. As a result, many researchers are focusing their efforts on replacing these toxic inhibitors with non-toxic ones. There is great interest in the use of green corrosion inhibitors, as they are non-toxic, environmentally friendly, readily available and renewable. They can be safely extracted by simple low-cost procedures from natural products such as plants that show a very high inhibitory efficacy [11-13].The plant extract are rich sources of molecules which have appreciably high inhibition efficiency and hence termed as "Green Inhibitors" [14]. These inhibitors are biodegradable and do not contain heavy metals or other toxic compounds [15]. The main constituents of the plant extracts have been reported to be a wide variety of organic compounds, including polyphenols, terpenes, carboxylic acids and alkaloids.Therefore, most of these compounds contain P, N, S, O atoms and multiple bonds in their structure, which serve as bonding centres for their adsorption on the surface of the zinc materials.

There are three types of inhibitors according to mechanism of electrochemical action, such as anodic, cathodic or mixed inhibitors. Anodic inhibitors slow down the oxidation reaction, i.e. by blocking the anodic sites (seat of metal oxidation), which decrease the density of the metaldissolution current and shift the corrosion potential in the positive direction. Cathodic inhibitors show blocking the cathodic sites (oxygen reduction site in aerated neutral environment or H^+ proton reduction site inacidic environment) which decrease the density of the hydrogen reduction current and which displace the corrosion potential in the negative direction. Cathodic inhibitors are considered safer

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than anodic inhibitors because they are not likely to promote localized corrosion. Third type of inhibitors is mixed inhibitors which act on both cathodic and anodic reactions. They decrease the speed of both partial reactions with little change in the corrosion potential. Corrosion can be slowed down by adsorption of an inhibitor on the metal surface.

The adsorption isotherm can be expressed as the relationship between the rate of recovery of an interface by the adsorbed species and the concentration of the species in solution [16,17]. An adsorption isotherm very much depends on the nature of metal, environment and amino acids used. There are several models of adsorption isotherms, we can cite the four most frequently used models such as Langmuir isotherm, Temkin isotherm, Freundlich isotherm and Flory-Huggins adsorption isotherm explain the conditions necessary for their validity in each case.

Langmuir Isotherm

This model assumes that there is a fixed number of sites on the surface of the metal, each site can only adsorb a single particle, so the adsorption energy is constant. The rate of adsorption is proportional to the inhibitor concentration and the number of unoccupied sites.

Temkin Isotherm

For this type of isotherm, the standard free adsorption energy decreases linearly with the recovery rate. It is also assumed that there are interactions (attraction or repulsion) between the adsorbate and the adsorbent, resulting in multilayer coating [18]. The Temkin isotherm is valid only for an intermediate range of ion concentrations [19].

Freundlich isotherm

Freundlich isotherm is applicable to adsorption processes that occur on heterogonous surfaces. This isotherm gives an expression which defines the surface heterogeneity and the exponential distribution of active sites and their energies [20].

Flory-Huggins adsorption isotherm

The Flory-Huggins model is sometimes used for interpretation of adsorption process of corrosion inhibitor molecules.

Medium

In this overview, plant extracts are used for controlling the corrosion of zinc in various medium has been studied. This research is mainly focused on HCl medium [21,22,25,28,30-32,34,37,39,40,42,43,46,47,49,50,52,54] and H_2SO_4[23,24,26,32,44,45,48,53]. But few mediums like NaCl [35,37,38,41,51,56], NaOH [34,37], Natural Sea water [27,29,33,36] and CH_3COOH [55]were also used.

Plant materials

Different parts of the plant such as leaves [26,29,36,40,44,47-50,54], fruits [52], seeds [32,55], peel [27]and plant extract [21-25,28,30,31,33-35,37-39,41-43,45,46,51,53,56] were used as corrosion inhibitors.

Extracts

Since it is generally known that the yield of chemical extraction depends on the type of solvent with varying polarities, extraction time, and temperature, the mass to-solvent ratio, and the chemical composition and physical characteristics of the samples, much attention is devoted to the choice of the most suitable organic solvent.Various solvents like ethyl alcohol [24,28,29,31,40,41,47,48,52,54], methyl alcohol [35,42,43,51], alcohol [23,27,36] , acetone [49,50] , distilled water [21,22,26,30,32,37,39,44,45,55,56] were used to prepare extract of plant materials. No indication of what extracts was used[25,33,34,38,46,53].

Additives

Various inhibitors have been used as corrosion inhibitor alone or combination with additive such as KI [32,40], KCl[40], Na_2S [35,51] and 1,5 Diphenyl carbazone [43] were used in combination with green inhibitor to study their synergistic effect meaning that the corrosion inhibition performance is higher for the mixture than for the individual components alone.

Methods

Different methods have been used to determine the inhibition efficiency of different inhibitors byWeight loss [21-32,34-36,39,43-55], Temperature [23,26-28,34,39,49,50,52], Thermometric [39-41,44,45,47,48], Gasometric[54], Hydrogen evolution [21-23,46,53], Potentiodynamic polarization [21,22,30,32,33,35,37-39, 42,46,51,54-56],Electrochemical Impedance Spectroscopy(EIS)[22,30,32,33,35,38,39,51,55,56], Synergistic effect [32,35,37,40,43,51] , pH [55] and Kinetic parameters: Rare constant and Half- life period [43] and Cyclic-Voltameter[42].

Metal surface analysis

When plant extracts were added in corrosive solutions to prevent the corrosion of metal a protective films formed on metal surface which is confirmed by different examination techniques, such as Scanning Electron Microscope (SEM) [28,29,32,33,36,51,52], Atomic Force Microscope (AFM) [33,51], Fourier Transform InfraRed (FT-IR) [28,29,31,36,47,48,52], UV spectra [28,29,36,52], Energy Dispersive X-ray (EDX) technique [28,29,36] , X-Ray

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Diffraction (XRD) [28,29,31,52] , XPS [32], IR spectroscopy [25,42] and Electrochemical frequency Modulation (EFM) [22,35,51].

Temperature effect

Room temperature [26-32,35,36,39,43-55] and higher temperatures [26-30,32,34,39,49-52,55] were used for the study of inhibition efficiency.

Adsorption isotherms

The adsorption behaviour of different inhibitors on the zinc surface has been investigated. The following adsorption isotherms have been obeyedsuch as Langmuir adsorption isotherm [21,25, 27-33, 35-37,39-41,43,46-48, 51-56], Freundlich adsorption isotherm [51], Temkin adsorption isotherm [22,23,27-29,52] and Flory-Huggins adsorption isotherm [56]. A list of various plant extract used as green corrosion inhibitors for zinc was shown in Table-1.

Table 1: Green corrosion inhibitors for Zinc in different media.

No.	Inhibitor	Medium	Methods	Findings	I.E. (%)	Year	Ref.
1	*Achillea fragrantissima*	HCl	weight loss, hydrogen evolution and polarization measurements.	Langmuir adsorption isotherm.	82.0 wl 70.2 pp	Ali et. al., 2014	21
2	*Ailanthus altissima* extract	HCl	weight loss, hydrogen evolution (HE), potentiodynamic polarization (PP), electrochemical impedance spectroscopy (EIS) and electrochemical frequency modulation (EFM) techniques.	mixed-type inhibitor, Temkin isotherm.	77.5 wl 76.9 pp	Fouda et al., 2018	22
3	*Allium Sativum*extract	0.5 N H_2SO_4	Weightloss, gasometry, thermometry.	Temkin adsorption isotherm.	81.20 wl	Pasupathy et al., 2014	23
4	*Allium Cepa* extract	2.0 M H_2SO_4	Weight loss with time.	I.E. is proportional to the volume of inhibitor and inversely proportional to time.	89.10 wl	Chinweuba, 2014	24
5	*Aloe vera*	2 M	Weight loss,	Langmuir	-	Aboia	25

		HCl	Infrared spectrophotometer.	adsorption isotherm, A first-order kinetics relationship		and James, 2010	
6	*Azadirachta indica* Leaves	H_2SO_4	Gravimetric with temperature.	I.E. decreases with increase in temperature.	83.58 wl	Sharma et al., 2009a	26
7	*Citrullus Vulgaris* Peel	Natural Sea Water	weight loss with time and temperature. FTIR and XRD studies.	Langmuir and Temkin adsorption isotherm.	82.81 wl	Petchiam-mal et al., 2012	27
8	*CnidoscolusChayamansa*	2 N HCl	Weight loss with time and temperature. UV, FT-IR, XRD and SEM-EDX techniques.	Langmuir and Temkin adsorption isotherm.	72.90 wl	Bright et al., 2015	28
9	*CnidoscolusChayamansa*Leaves extract	Natural Sea Water	Weight loss with temperature, UV, FT-IR, XRD and SEM-EDX techniques.	Langmuir and Temkin adsorption isotherm.	87.73 wl	Bright et al., 2015	29
10	*Elettariacardamomum* extract	2.0 M HCl	weight loss, potentiodynamic polarization, and electrochemical impedance spectroscopy.	mixed-type inhibitor, Langmuir adsorption isotherm.	93.6 wl 93.8pp 89.2EIS	Sobhi 2012	30
11	*Eugenia jambolana*	2.0 N	Weight loss, UV, FT-IR and XRD	Langmuir adsorption	86.56 wl	Deepa Rani	31

	extract	HCl	spectral studies.	isotherm.		et al., 2013	
12	Fenugreek seeds extract	HCl-H_2SO_4 + KI	Weight loss and Potentiodynamic polarization and EIS measurements Scanning electron microscope (SEM).X-rays photoelectron spectroscopy (XPS) analysis.	Mixed-type inhibitor, Langmuir adsorption Isotherms.	90.70 wl in H_2SO_4 and 66.60 wl in HCl . 64.83 pp HCl 65.35 EIS	Shimaa and Hamedh, 2016	32
13	Fucoidan	Natural Sea water	Potentiodynamic polarization and EIS measurements. Scanning electron microscope (SEM). X-rays and atomic force microscope (AFM) analysis. .	Anodic inhibitor, Langmuir adsorption Isotherms.	---	Wang et al., 2017	33
14	*Hibiscus subdariffa* (karkade) extract	HCl and NaOH	Weight loss, thermometric experiments.	--	85.0 wl	Hosary et al., 1972	34
15	Khillah Extract	3.5 % NaCl + 16 ppm Na_2S	weight loss, potentiodynamic polarization, electrochemical impedance spectroscopy (EIS) and electrochemical frequency modulation (EFM) techniques.	Mixed-type inhibitor, Langmuir adsorption isotherm.	87.2 wl 86.8 pp	Fouda et al., 2015	35
16	*Kingiodendron pinnatum* leaves (KPL)	Natural Sea Water	Weight loss method, UV, FT-IR and SEM-EDX Spectral techniques.	Langmuir adsorption isotherm.	92.1 wl	Deivanay-agam et al., 2015	36

17	Lawsonia extract	0.1 M HCl+ 3.5% NaCl+ 0.1 M NaOH	polarization technique.	Mixed-type inhibitor, Langmuir adsorption .	In HCl-76.19 pp, 93.44 pp in NaCl and 76.92 pp in NaOH	El-Etre et al., 2015	37
18	*Mansoaalliacea* plant extract	3 % NaCl	Potentiodynamicpolarization and electrochemical impedance spectroscopy (EIS).	mixed-type inhibitor. .	90 .0 pp	Suedile et al., 2014	38
19	Marjoram	1.0 M HCl	Weight loss with temperature, thermometric, polarization and EIS techniques.	Mixed-type inhibitor. Langmuir adsorption isotherm	91.8wl, 92.5 pp 88.8 EIS	Sobhi Et al., 2013	39
20	*Moringa oleifera* Leaves	HCl +KCl and KI	Thermometric Method. Synergistic effect	Langmuir adsorption isotherm.	91.3 wl	Ugbe et al, 2015-14	40
21	*Moringa oleifera*	polluted NaCl	Thermometric Method.	Langmuir adsorption isotherm.	86.0 wl	Fouda et al., 2014	41
22	*Nictanthes* plant extract	HCl	Potentiometry polarization and Cyclic-Voltameter, IR spectra.	Rate of corrosion is reduced with the increasing concentration of inhibitor.	--	Chauhan et al., 2013	42
23	*NypafruticansWurmb* extract	HCl + 1,5 Diph EnylCarba zone.	weight loss with time, Rate constant and Half-life.	Langmuir and Temkin adsorption isotherms.	--	OrubiteOkorosaye and Oforka, 2004.	43
24	*Ocimumtenuiflorum*(Tulasi)	H_2SO_4	gravimetric and thermometric	chemical adsorption	77.61 wl	Venkata Naga	44

	Leaves extract		techniques.	,		BajiTokala, 2018	
25	*Ocimumtenuiflorum* (Tulsi)	H_2SO_4	gravimetric and thermometric techniques.	--	86.25 wl	Sharma et al., 2009	45
26	Onion juice	HCl	weight loss and hydrogen evolution measurements. potentiodynamic polarization technique.	Mixed-type inhibitor. Langmuir adsorption isotherm. spontaneity of the adsorption process.	84.74 wl	El-Etre, 2006	46
27	Picralimanitida Leaves	1.0 M HCl	weight loss and thermometric methods. FTIR analysis.	Langmuir adsorption isotherm.	86.78 wl	Ezeugo et al., 2018	47
28	Picralimanitida leaf,	0.5 M H_2SO_4	weight loss, and thermometry methods. Fourier transform infrared (FTIR) analysis.	Langmuir adsorption isotherm.	84.95 wl	Onukwuli and Ezeugo, 2018	48
29	Red onion skin extract (leaves of Aloe vera)	2.0 M HCl	weight loss with temperature.	--	--	James and Akaranta, 2009	49
30	Red onion skin extract	HCl	weight loss with time and temperature.	--	70.0 wl	James and Akaranta, 2011	50
31	*Slanum Nigrum* Extract	3.5% NaCl and 16 ppm Na_2S	Weight loss with temperature, Tafel polarization, electrochemical impedance spectroscopy (EIS), SEM, AFM and electrochemical frequency modulation (EFM) techniques.	Mixed-type Inhibitor. Langmuir and Freundlich adsorption isotherms.	81.5wl, 81.8pp 81.5EIS.	Fouda et al., 2017	51

32	*Solanum Torvum* fruits extract	1.0 N HCl	Weight loss measurements with various time and temperature. UV, FT-IR, XRD and SEM-EDX techniques.	Langmuir and Temkin adsorption isotherm.	80.90 wl	Bright et al., 2015	52
33	*Streptomyces griseus* (Albomycin)	H_2SO_4	weight loss and hydrogen evolution methods.	Langmuir adsorption Isotherm. spontaneous, exothermic,	82.68 wl	Nnabuk Eddy Okon, 2010	53
34	*Vernonia Amygdalina* (Bitter Leaf)	2 M HCl	weight loss, gasometric and potentiostatic polarization, Atomic Force Microscopy (AFM)	Langmuir adsorption Isotherm.	93.00 wl	Popoola and Fayomi, 2011	54
35	*VulgarisThymus* (VT), *CuminumCyminum*(CC), *Hibiscus Subdariffa*(HS), and *Feungreek*Seeds (FS)	2 M CH_3-COOH	weight loss with time and temperature, pH and electrochemical measurements.	Mixed-type inhibitors. Langmuir adsorption isotherm.	--	El- Aila et al., 2010	55
36	Lupine, Hlfabar and Damssisa extracts	0.5M NaCl	potentiodynamic polarization and electrochemical impedance spectroscopy (EIS) techniques.	Mixed-type inhibitors. Langmuir and Flory-Huggins adsorption isotherm.	89.10, 94.70 and 90.70 EIS for lupine, hlfabar and damsses a	Abd-El-Naby et al., 2012	56

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					extracts respectively		

[The inhibition efficiency is calculated according to the data achieved by weight loss (Wl) measurements, potentiodynamic polarization (PP), and electrochemical impedance spectroscopy (EIS) measurements.]

phytochemical constituents

Plant extraction products are acts as a good potential corrosion inhibitor for zinc in various acidic, alkaline and other media. The active constituents of green inhibitors are varied from one plant species to another species but their structures are closely related to their number of organic molecules, e.g.,***Achillea fragrantissima***[21] contain α-Thujone (60.9%), β-Thujone (9.1%), sabinene (4.1%) and camphor (3.7%) were characterized as the main constitutes. ***Ailanthus altissima***[22,57]contain of alkaloids, terpenoids, steroids, and flavonoids. ***Allium sativum***[23] contain alliin, allicin, dially sulphide and ally methyl trisulphide. ***Aloe vera***[25, 58] contain compounds such as salicylates, magnesium lactate, acemennan, lupeol, campestrol, sterol, linolenic, aloctin and anthraquinones. ***Azadirachtaindica*** **(Neem)** [26]contain Nimbolinin, Nimbin, Nimbidin, Nimbidol ,Salannin, alkaloids, fatty acids, and nitrogen and oxygen-containing compounds. ***Citrullus Vulgaris*** **peel**[27] contain Citrulline as main active phytoconstituents. ***CnidoscolusChayamansa*** **(Chaya)**[28,29] leaves are very high in protein, calcium, iron, carotene, and vitamins A, B and C, also contains Flavonoids, Terpenoids, Glycosides, Steroids, Alkaloids, Carbohydrates, Amino acid, Tannins, Chlorides, Copper, Nitrates, Potassium, Zinc, Carbonates and bicarbonates. ***Elettariacardamomum***[30, 59]The main compound is 1,8-cineole (representing 50 % or more), with smaller amounts of α-terpineol and limonene. ***Eugenia Jambolana***[31,60] fruit peel contains anthocyanins, delphinidin, petunidin, malvidindiglucosides. **Fenugreek plant**[32,61] contain active constituents like alkaloids, amino acids, saponins, seroidalsapinogens, fiber, coumarin, lipids, vitamins and minerals.***Hibiscus Subdariffa*** **(karkade)**[34, 62] consists of proteins, glucides, organic acids (citric, malic, and traces of tartaric acid). **Khillah**[35, 63] contains small amount of volatile fatty oil, proteins 13.83 %, cellulose 22.4 % coumarins 0.5 % and furanochromones up to 4 % such as khellin 1.2 % and visnagin 0.3 %. ***Kingiodendronpinnatum*** (KPL)[36] contain flavonoids, alkaloids, saponins and triterpenes. ***Lawsoniainermis*** **(Henna)** [37] contains naphthoquinone, Phenolic compounds, Terpenoids, Sterols, Coumarin, Amino acid and Fatty acid..**Marjoram** (**Origanummarjorana L.)** oil extract [39,64] mainly contain thymol (38.4%) and cis-sabinene hydrate (25.3%). Other components detected in lower amounts in oil sample were sabinene and p-cymene (up to 7.4% and 13.9% in), and alpha-terpinene (up to 13.3%) in addition to some active phytochemicals like terpines, flavonoids, and rosmarinic acid is found in big quantities. ***Moringa oleifera***[40,41]leaves contain phytochemicals such as saponins, tannins, flavonoids, glycosides, carbohydrates, reducing sugars, terpenoids, steroids and alkaloids.***Ocimumtenuiflorum*** **(Tulasi)**[44,45] mainly grouped as essential oils (0.516-0.596%), total carbohydrates (11.87-11.50%), total flavonoids (0.735-0.945%) and proline (0.28-0.536 mmol/g leaves fresh weight). Omer et al. [65] further determined that the essential oil of O. tenuiflorum contained higher amount of linalool (39.39-55.26%) and moderate amounts of methyl chavicol (0-6.66%), nerol (0.48-8.0%), geraniol (0.26-1.75%) and citral (4.26-6.79%). **Onion juice**[46, 66] contains proteins, lipids, carbohydrates and fibers. The juice is characterized by the presence of two sulfur-containing amino acids (glutamyl peptides), namely S-(1-propenyl)-L- cysteine sulfoxide and S-2-carboxypropyl glutathione. **Picralimanitida leaf**[47,48] contains many organic compounds, such as phenolics, terpenoids, and tannins as their major phytocompounds and also saponins, flavonoids, tannins and steroids and alkaloids in moderate amount to scavenge free radicals induce detoxification. **Red onion skin**[49,50] mainly contains Quercetin. It is a compound with conjugated system and contains hetero atoms and carbonyl groups that are electron rich. ***Solanum Torvum***[52] contain phytoconstituents such as calcium, iron, carbohydrate, thiamine, niacin, solasodine, flavonoids, alkaloids, phenolic compounds, saponins, terpenoids, tannins etc. ***Streptomyces griseus*** **(Albomycin)**[53], ***Vernonia Amygdalina*****(Bitter Leaf)** [54]leaf extract contains alkaloid, saponins, flavonoids, tannins etc. ***Cuminumcyminum*** **(Cumin, Jeera)** [55] contain Cuminaldehyde can be predominantly affecting the oxygen atom present in it. **Lupine**[56] seeds contain up to 5% quinolizidine alkaloids, lupanine was the most abundant, multiflorine and sparteine, **Hlfabar**[56] contains hydroxyl-α-eudesmol derivatives. **Damssisa**[56] contain Lactones, damsin, ambrosin and coumarins.

II.CONCLUSION

The known hazardous effects of most synthetic organic inhibitors and restrictive environmental regulations are driving researchers to focus on the need to develop low-cost, non-toxic and environmentally friendly natural products as effective corrosion inhibitors for many metals and alloys in various aggressive environments. These natural organic compounds are either synthesized or extracted from aromatic plants, spices and medicinal plants. Plant extracts are considered to be an incredibly rich source of biosynthetic chemical compounds that can be extracted by simple, low-cost procedures and are water-soluble and biodegradable in nature.

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