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Review Paper

Heena (*Lawsonia inermis* L.) as a green inhibitor for prevention of metals and alloys from corrosion-A Review

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Abstract

Henna has been used as green corrosion inhibitor for prevention of various metals and alloys such as Al, MS, CS, SS, iron, zinc, copper, tin and brass in various media like acidic, basic or neutral. A percentage of I.E. of Heena was determined by using various techniques such as WL, PDP and EIS. Film occur on metal surface has been studied by using SEM, FT-IR, UV-Vis. spectroscopy and EDX techniques. Phsysorption of Henna extract on metal surface was found to obey various isotherms. PDP study reveals that Lawsonia inermis L. can function as mixed type or anodic or cathodic type of inhibitor.

Keywords: Corrosion, Inhibition, Heena, WL, PDP, SEM.

Introduction

Corrosion of metal/alloy, which can be defined as the deterioration of materials due to their reaction with the environment. Recent study indicates that corrosion causes economic losses of about 2.5 trillion US dollars a year, constituting almost 3.4% of the worldwide GDP[1]. Metals and alloys are exposed to hostile environments during their industrial usage, including manufacturing, processing and transportation, which accelerate their degradation. Most efficient methods for protection metals against corrosion is the use of inhibitors[2]. The application of the organic and inorganic inhibitors not only gives toxicity effect towards living organism, also it is quite expensive. Nowadays a natural products are used as corrosion inhibitors as they are eco-friendly, cheap, easily available, non-toxic and ecologically acceptable[3,4]. Plant extracts have the required inhibiting properties, i.e. they contain various organic compounds with heteroatoms like nitrogen, oxygen and conjugated bonds[5-7].

The organic molecules used as inhibitors, can be adsorbed on the surface of alloy through these heteroatoms and their conjugated system. This adsorption is either physical, by electrostatic interaction (physisorption) or chemical through covalent bond formation (chemisorption).

Lawsonia inermis L. (Henna) belongs to the family Lythreaceae and the sole member of its genus *Lawsonia*. It is glabrous and multi-branched shrub or small tree (2 to 6m in height). It is commonly known as henna in Arabic and mehndi in Hindi. It is a native to North Africa, Asia and Australia and it is cultivated in the tropics of America, Egypt, India and parts of the Middle East[8]. The leaves are small, sub-sessile and greenish brown to dull green in color. Flowers are white, small and numerous. Fruits are small, brown, globose capsules, petals orbicular to obovate, white or red. Seed capsules are red, globose, with numerous tiny pyramidal[9].

Traditional uses: In Ethiopia, *Lawsonia inermis* L is traditionally used to develop a red or black coloring to hands, skin, nails and hair in weddings and religious festivals[10]. The leaves are used in the treatments of wounds, ulcers, cough, bronchitis, lumbago, rheumatagia, inflammations, diarrhoea, dysentry, leucoderma, scabies, boils, anaemia, haemorrhages, fever, falling of hair and greyness hair[11,12]. Leaves of Heena also used in dysuria, bleeding disorder, prurigo and other obstinate skin diseases[13,14]. The bark is used as its antibacterial, antifungal, antihemorrhagic, hypotensive and sedative activity[15]. Several studies also indicates its various activities such as antimicrobial, antibacterial, antioxidant, anti-corrosion, anti-inflammatory, antipyretic, anti-tumoral activity[16].

Various techniques have been used to analyse the nature of protective film formed on the metal surface. Depending on the nature of metal and corrosive environment Heena obeys different types of isotherms and shows various type of inhibition, namely anodic, cathodic or mixed type. The main purpose of this review paper is to present the results regarding inhibition action of henna extract on some metals and alloys such as Al, MS, CS, Iron, zinc, tin and nickel in various acidic, basic and neutral solutions has been investigated.

Materials and Methods

Various metals have been used in corrosion inhibition study. Different methods were employed to determine the corrosion inhibition process.

Metals used: Various metals have been used in corrosion inhibition study such as Al [17-19,25], Al-Alloy-AA 5083[20-24], Brass [25], Copper [25-29], C.S.[30--34], Nickel[30], CS-Alloy [33], 304 S.S.[35], N 80 Steel[36], Steel[37], Steel-37[19], Iron[38], MS [39-50],Tin [51] and Zinc[30, 52].

Medium Usually corrosion of metals in various environments has been studied. For example acid medium[17-19,25,26,28-31,34-42,44,49-51], basic medium[19,25,30,43,51], Sea water[20-24,45,48], Well water[46,47], chloride medium[19,25,30,32,33], CCl_4 [27].

Plant parts: Various parts of Henna plants like Seed [17], Leaves [18,21,24-26, 28-30,35,36,39,40,44,46,48,50,51], Powder [31,32,39,42,43,45] were used for corrosion inhibition study.

Solvents: To prepare extract **s**olvents like Distilled water [26,27,33,36,39,48,50,51], Double distilled water [34,35,46, 52] and Ethanol [20, 21,24,29,40,45,49] were used.

Techniques: Different techniques, like WL with temperature [17,18,26,27,31,34,35,38,39,43,44,50,51, 52], WL with time [17,20-23,28,29,40,42,45,52], PDP [18,20,21,24-28,30-34,36,37,39,41,42,46,47,50,52] and EIS[18,20,24,26, 27,31,33,36,39,42,45-47,50] were employed to evaluate the corrosion I.E. of Henna.

Film analysis: Metal surface film were studied using different techniques, such as SEM [24,29,31,33-35,37,40-43], FT-IR spectroscopy [17,20,22-24,29,31,32,34,37,40,45,51],UV-Vis. Spectroscopy [17,29,40,43,46,47], EDX [17,31,33,34], EDS [41], AFM [46,47], XPS[37] , XRF [40] and GC-MS [50] have been employed.

Other techniques: QCC [21,40], OCP [28,33], QCA [29], FEM [31], SE [32], FS [32,46,47] were also used.

Adsorption isotherms: Various isotherms like Langmuir [18,21,22,24,26,27,29-31,34,38-40,43,45,50, 52], Temkin [17,19,25,27], Freundlich [27,52] and Flory–Huggins isotherm[37] were suggested.

Corrosion inhibition study of Heena extract on various metals and alloys in different media is presented in Table-1.

Table-1: Corrosion inhibition of various metals and alloys by Heena as an inhibitor.

Metal / Alloy	Medium + Additive	Techniques used	Findings	I.E. max. (in %)	Ref. No.
Al	0.5N HCl	WL with time & temperature, UV-Vis., EDX, FT-IR.	Temkin isotherm, adsorption process is exothermic, endothermic	84.52 WL	17
Al	0.75M HCl	WL with Temp., EIS, PDP.	Langmuir isotherm, Mixed-type of inhibitor	85.34 WL, 88.14 PDP, 86.63 EIS	18
Steel 37 and Al	0.1HCl, 0.1M NaOH, 0.1M NaCl	WL	Temkin adsorption isotherm,	96.0% for Steel-HCl 99.8% with Al-NaOH	19
Al Alloy AA 5083	Sea water	PDP, FT-IR, EIS, WL with time.	Mixed inhibitor and predominantly cathodic in nature	---	20
Al Alloy AA 5083	Tropical Sea water	WL with time, EIS, PDP, QCC.	Mixed type of inhibitor. Langmuir isotherm	93.0 WL, 95.0 PDP, 94.0 EIS	21
Al Alloy AA 5083	Sea water	WL with time, FT-IR, EIS.	Langmuir isotherm.	88.0 EIS	22
Al Alloy AA 5083	Sea water	WL with time, EIS, FT-IR.	---	92.61 EIS	23
Al Alloy AA 5083	Sea water	EIS, PDP, SEM, FT-IR.	Mixed -type inhibitor. Langmuir adsorption isotherm	93.0 PDP, 92.0 EIS	24
Al, Copper and Brass	NaOH, acid chloride	WL, PDP.	Temkin adsorption isotherm.	--	25
Copper	0.5, 0.75 and 1M HNO_3	WL with temperature, PDP, EIS.	Mixed -type of inhibitor. Langmuir isotherm.	89.76 WL, 89.36 PDP, 84.23 EIS	26
Copper	CCl_4	WL with temperature, PDP, EIS.	Temkin, Freundlich and Langmuir adsorption isotherm.	83.0 WL, 93.83 PDP	27
Copper	0.5 M HCl	WL with time, OCP, PDP.	----	58.7 WL, 54.0 PDP	28
Copper	0.5 M HCl	WL with time, FT-IR, SEM, UV-Vis., QCA.	Langmuir adsorption isotherm.	90.01 WL	29
C-Steel, Nickel and Zn	0.1M HCl + 3.5% NaCl + 0.1M NaOH	PDP.	Mixed-type inhibitor, Langmuir adsorption.	HCl 95.78 PDP for C-Steel. NaCl 93.44 PDP for Zn, NaOH 76.92 PDP for Zn.	30

CS	1.0 M HCl	WL with temperature, PDP, EIS, EDX, FEM, FT-IR, SEM.	Mixed-type of inhibitor. Langmuir isotherm.	81.5 WL, 83.1 PDP, 79.5 EIS	31
CS	60 ppm of Cl^- solution.	WL, PDP, FT-IR, SE, FS	Cathodic type of inhibitor.	81.0 WL with Heena, 98.0 WL with Henna+Zn^{+2}	32
CS- Alloy	3.5% NaCl	PDP, EIS, OCP, SEM, EDX.	Mixed -type inhibitor but more anodic inhibitor.	81.68 PDP	33
CS	1.0 M HCl	PDP, EDX, SEM, FT-IR, WL with temperature.	Langmuir isotherm. Mixed -type inhibitor.	88.42 WL, 92.72 PDP	34
304 SS	1.0 M H_2SO_4	WL with temperature, SEM	Efficient inhibitor	96.2 WL	35
N 80 Steel	Mud Acid HCl/HF12/3% Wt.	WL, PDP, EIS.	Cathodic type inhibitor.	84.21 WL, 92.62 PDP, 81.11 EIS	36
Steel	8 M H_3PO_4	PDP, SEM, FT-IR, XPS.	Anodic -type inhibitor. Flory–Huggins isotherm.	67.6 PDP	37
Iron	1.0 M HCl	WL with temperature.	Langmuir adsorption isotherm.	--	38
MS	0.5- 2.0 M Acetic acid	WL with temp., PDP, EIS.	Mixed -type of inhibitor. Langmuir adsorption isotherm.	86.36 WL, 80.91 PDP, 95.98 EIS	39
MS	0.5 M HCl	WL with time, SEM, FT-IR, UV-Vis., QCC, XRF.	Mixed -type of inhibitor. Langmuir adsorption isotherm.	88.26 WL	40
MS	1.0 M HCl	PDP, SEM, EDS.	Mixed-type of inhibitor.	92.6 PDP	41
MS	5% HCl	WL with time, PDP, EIS, SEM.	---	99.2 PDP	42
MS	1M NaOH+ Henna /Zeolite powder	WL with temperature, SEM, UV spectra.	Langmuir adsorption isotherm.	95.97 WL	43
MS	1 N H_2SO_4	WL with temperature.	---	80.68 WL	44
MS	Sea water	WL with time, EIS, FT-IR.	Anodic-type inhibitor.	--	45
MS	Well water	WL, PDP, EIS, UV-Vis., AFM, FS.	Mixed -type inhibitor.	96.0 WL, 72.4 PDP, 78.8 EIS	46
MS	Well water	WL, PDP, EIS, UV-Vis., AFM, FS.	Mixed -type inhibitor.	96.0 WL, 72.05 PDP, 78.79 EIS	47
MS	Synthetic Sea water + H_2S	WL	---	75.O WL	48
MS	1.0 M HCl	WL	Langmuir adsorption isotherm.	93.14 WL	49
MS	1.0 M HCl & 0.5 M Acetic acid	PDP, EIS, GC-MS, WL with temperature.	Mixed-type of inhibitor. Langmuir adsorption isotherm.	HCl 92.59 WL, 94.44 PDP, 90.34 EIS. Acetic acid 86.27 WL, 80.91 PDP, 95.98 EIS.	50
Tin	1.0 M HCl & 1.0 M NaOH	WL with temperature, FT-IR.	Freundlich isotherm.	HCl 85.19 WL, NaOH 95.45 WL	51
Zn	2.0 M Acetic acid	WL with temperature and time, PDP.	Mixed-type of inhibitor. Langmuir isotherm	76.0 WL	52

Abbreviations: AFM: atomic force microscopy, Al: aluminum, CS: carbon steel, EDX: energy-dispersive X-ray spectroscopy, EIS: electrochemical impedance spectroscopy, EDS: energy-dispersive spectroscopy, FS: fluorescence spectra, FT-IR: fourier-transform infrared spectroscopy, FEM: electrochemical frequency modulation, I.E.: inhibition efficiency, IE max (%): maximum inhibition efficiency, MS: mild steel, NaOH: sodium hydroxide, OCP: open circuit potential, PDP: potentiodynamic polarization, SE: synergistic effect, SEM: scanning electron microscopy, SS: stainless steel, PPM: parts per million, QCA: quantum chemical ananlysis, QCC: quantum chemical calculations, , UV-vis.: ultraviolet-visible spectroscopy , WL: weight loss, XPS: X-ray photoelectron spectroscopy, XRF: X-ray fluorescence, Zn: zinc.

FT-IR Study: The FT-IR spectrum of Laws oniainermis extract is shown in Figure-1to exhibit a functional groups and bonds such as O–H,=C–H, C=C, C=O etc.[37].

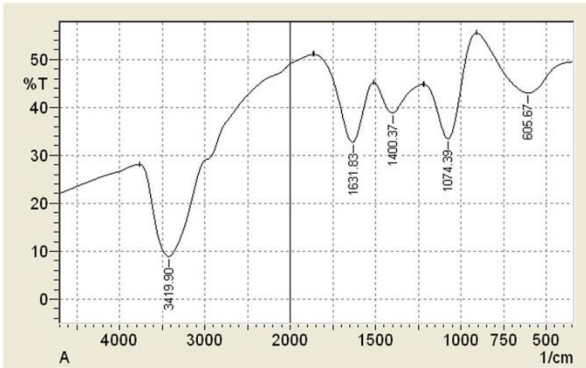


Figure-1: FTIR spectrum of *Lawsonia inermis*.

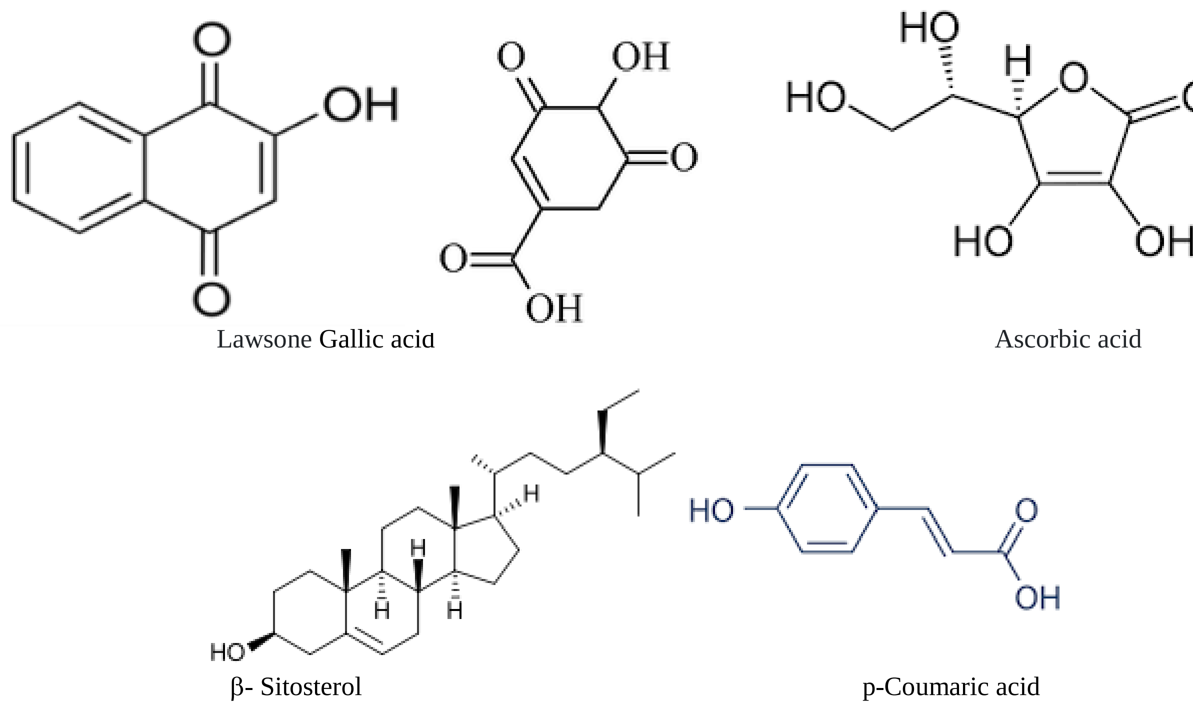
Chemical composition of Henna leaves: The main constitutes of Heena extract are hydroxy aromatic compounds which consists tannin and Lawsone[30,41,53]. Lawsone contained at 1.0 to 1.4% in the leaves of Henna[54]. Other compounds, such as triterpenoids, steroids (β-sitosterol)[9], carbohydrates, phenolic compounds, flavonoids (luteolins, apigenin, and their glycosides), saponins, proteins, alkaloids, terpenoids, quinones, coumarins, xanthones, fatty acids, resin and tannins[17,30,41,49, 52,55-58]. Some research shows that henna leaves contain soluble matter, Lawsone, gallic acid and dextrose[41,50,59]. Tannin or also known as tannic acid present with a core glucose and methyl gallate groups[30]. Some of the major active constituents of Heena shown in Figure-2[29,40].

Mechanism of corrosion inhibition

Mechanism of corrosion inhibition depends on the adsorption of inhibitor on metal surface. Adsorption process is influenced by the nature of metal and chemical structure of the inhibitor[41]. Main constitutes of Heena are tannin and Laws one. Tannins are also known to form complex compounds with different metal cations, especially in the basic media. The inhibitive action of tannin was attributed to the formation of a passivating layer of tannates on the metal surface[30,60,61]. The main constituent of Henna extract is Lawsone. The structure of lawsone ($C_{10}H_6O_3$) shown in Figure-2[26,39], which is consists of benzene unit, p-benzoquinone unit and phenolic group[36].

Lawsone molecule is a ligand that can chelate with various metal cations forming insoluble complex compounds adsorbed on the metal surface[31] is a probable interpretation of the observed inhibition action of lawsone[36,41,50]. The process of adsorption is depends on molecular structure, solutions composition, nature of the metal surface and temperature[22].

In acidic medium, delocalization of the lone pair of electrons on hydroxyl group takes place causing rearrangement of lawasone structure as shown in Figure-3[18,26,39,62].



Lawsone Gallic acid Ascorbic acid

β- Sitosterol p-Coumaric acid

Figure-2: Major active constituents of *Lawsonia inermis*.

The migration of the hydrogen atom with a pair of electrons from an adjacent carbon to the carbon bearing the positive charge. This arrangement is intra-molecular and the reaction product are structural isomers (Figure-3). Such a rearrangement, in the presence of metal cations, enhance the complex formation reaction shown in Figure-4[18,49,59]. This could be the reason for the observed high I.E. in acidic medium[36,41].

Tannins are found limited inhibition in acidic media than in alkaline medium[30]. It can be due to the higher amount of lawsone in Heena leaves extract.

SEM Tests: The SEM micrographs for CS exposed in 1M HCl in absence and presence of 300ppm Henna were shown in Figure-5[31]. It was observed from Figure-5 that CS suffers from corrosion in 1M HCl and corrosion products appear very uneven shown in Blank SEM image. While in the presence of henna extract reduces the damage of CS surface and makes the surface smoother[31].

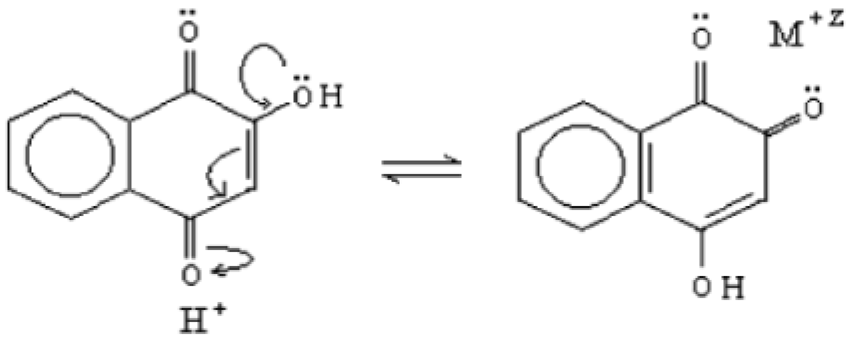


Figure-3: Electron delocalization process on the Lawsone molecule[18,41].

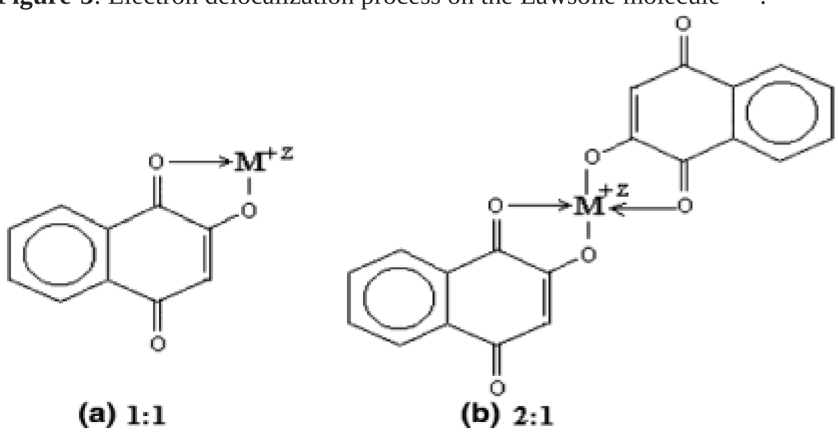


Figure-4: Forms of metal ion-lawsone complexes.

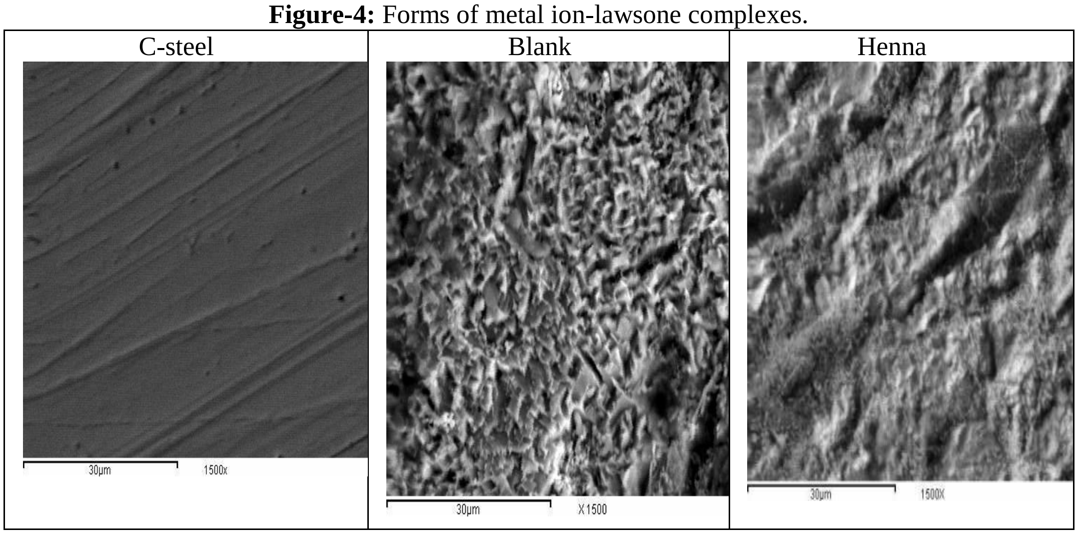


Figure-5: SEM micrographs for CS in 1 M HCl in presence and absence of 300 ppm Henna.

Conclusion

The present review paper summarized the research works carried out by various researchers on corrosion inhibition of various metals and alloys by using Henna as an eco-friendly inhibitor. Henna extract behaved as anodic, cathodic and mixed-type of inhibitor. Henna extract shows percentage I.E. in a range from 54.0 to 99.8%.

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