Int. J. Electrochem. Sci., 14 **(2019)** 4668 – 4682, doi: 10.20964/2019.05.47

International Journal of ELECTROCHEMICAL SCIENCE
www.electrochemsci.org

Henna Extract as Green Corrosion Inhibitor for Carbon Steel in Hydrochloric Acid Solution

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Received: 3 Janaury 2019 / *Accepted:* 18 February 2019 / *Published:* 10 April 2019

The extract of henna (*Lawsonia Inermis*) leaves have been evaluated as a green corrosion inhibitor for carbon steel (C-steel) in 1M HCl solution, utilizing mass loss (ML), potentiodynamic polarization (PP), AC impedance spectra (EIS) and electrochemical frequency modulation (FEM) techniques. The surface analysis was performed using energy dispersion spectroscopy (EDX) and scanning electron microscopy (SEM) methods. The inhibiting property of the extract is familiar with the presence of Lawsonia *as* a major constituent in the extract. PP study revealed that the adsorption of the extract affects both anodic and cathodic reactions. The inhibitor efficiency (IE %) was found to improve with rising the extract concentration and reached 83.1% at 300 ppm. On the other hand, the PE% was lowered by raising the temperature. The adsorbed Henna molecules on C-steel obeys Langmuir isotherm. The results achieved by different tests are close to each other.

Keywords: Corrosion, Henna leaves extract, HCl, C-steel and Green inhibitor

1. INTRODUCTION

Inhibitors are chemical compounds commonly utilized in minor concentrations whenever metal is in interaction with a corrosive solution. Carbon steel alloys are cost and have good mechanical properties paralleled to other metal alloys. On the other hand, it usually undergoes severe corrosion in altered functioning systems. There are altered forms of corrosion with altered mechanisms reliant on numerous factors such as the kind of metal, effective temperature, and environment. Therefore, the deep research is always wanted to hindrance corrosion by utilized inhibitors. The presence of such composite retards the corrosion procedure and preserves its rate to a smaller and thus inhibits economic damages owing to metallic corrosion. One of the sources of these clean and low-cost inhibitors is plant extracts.

Plant parts enclose numerous composites that content the mentioned criteria. Recently, scientists researches have adopted and attention for the use of naturally happening constituents for various metals and in acidic medium such as: Opuntia extract [1], Begonia extract [2], Prosopis juliflora plant extract [3], Artemisia Judaica Herbs extract [4], Aqueous Extract of Juniperus [5], Calotrpics procure plant extract [6], Tilia leafs [7], Cupressus sempervirens extract [8], aqueous extract of Coriander Seeds [9], Fennel seed extract [10], Rumex Vesicarius Extract [11], aqueous extract of Propolis [12]. The previous studies revealed that these plant extracts enclose many organic composites that contain heteroatoms like O, S, and N which act as adsorption centers.

The scope of the present work is to examine the (PE) of henna extract (*Lawsonia Inermis*) as a green inhibitor for C-steel corrosion utilizing different techniques.

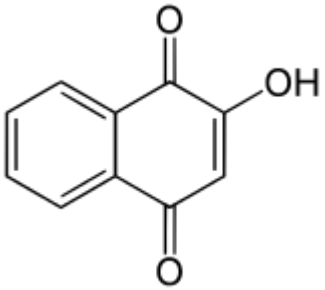
2. EXPERIMENTAL

2.1 Materials

The test specimens had cut from C-steel sheet of 0.02 cm in thickness and its composition is (weight %) is: 0.200 C, 0.340 Mn, 0.003 S, 0.022 P, and Fe balance. The all specimens were prepared, degreased and cleaned as described earlier [13]. Electrochemical tests were obtained utilizing a three compartment electrode cell. $Hg/Hg_2Cl_2/Cl^-$ (SCE) via a Luggin capillary probe was utilized as an electrode reference while the counter electrode is the PT foil and C-steel as the working electrode. All chemicals utilized (HCl, 37%) for this research was analytical grade and were utilized as established. The specimens were first dipped in the investigative solution for 25 minutes to attain a stable state [14]

2.2. Henna Extract Preparation

The Henna powered was obtained from the market and extracted for 4 h in boiling water. Then filtered after cooling, the evaporated the water from the *Henna* extract. The obtained solid residue by this procedure was again liquefied in 1000 mL bi-distilled (1000 ppm). The main component in Henna extract is:



Lawsone (2 - Hydroxy- 1, 4- naphthoquinone)

2.3. Mass Loss (ML) Test

ML test is a simple and reliable test utilized to measure the rate of corrosion rate (CR) of metals and %IE. After that, the samples were dipped in 1.0 M HCl presence and absence of adding an various concentration of *Henna extract*. After the dipping period gone, the coins were removed from the solutions, washed with bi-distilled water, dried and weight. MR tests were accepted at an altered temperature (298 to 328K) and repetitive three times to confirm the reproducibility of the results. The (IE) of the inhibitor and (CR) were measured utilizing Eq. (1) and (2) correspondingly [13]:

$$\% IE = [1–(W_1/W_2)] x100 \quad (1)$$

$$CR = [W_2–W_1]/At \quad (2)$$

where W_1 and W_2 are the ML (mg cm^{-2}) with and without the *Henna extract*, correspondingly, t is the period of dipping in min and A is the area of the coins in cm^{-2}.

2.5. Electrochemical Technique

Potentiodynamic polarization method was taken in a typical three compartment glass cell. The potential range was (-0.5 to 0.5 V vs. SCE) at OCP with a scan rate 1 mVs^{-1}. The PE was calculated using Eq. (3):

$$\% IE = [1- (i_{corr(inh)} / i_{corr(free)})] \times 100 \quad (3)$$

Where $i_{corr(free)}$ and $i_{corr(inh)}$ are the currents without and with *Henna extract*, correspondingly.

Impedance measurements were achieved by AC signs of 5 mV signal-to-signals amplitude and at a range of frequency among100 k Hz and 0.2 Hz. The % PE and the (θ) of the utilized Henna extract gotten from the EIS tests can be measured utilized Eq. (4):

$$\%IE = \theta \times 100 = [1- (R°_{ct}/R_{ct})] \times 100 \quad (4)$$

Where R^o_{ct} and R_{ct} are the resistances in the absence and presence of Henna extract, correspondingly.

EFM methods were conducted at two frequencies (2-5) Hz. The base frequency was 0.1 Hz. The (i_{corr}), (β_c and β_a) and (CF-2, CF-3) (Causality factors) was measured by the greater two peaks [14-15].

(PP), (EFM) and (EIS) techniques were performed utilizing the similar manner as earlier with a Gamry framework system depends on ESA400. Gamry apparatus contains DC105 software, EIS300 software for EIS and EFM140 software for EFM tests; the computer has utilized for calculating the data.

2.6. Surface Analysis

SEM and EDX were utilized for morphological investigation on the unprotected and protected C-steel coins after dipping for 24 h in test solutions in the presence and absence of 300 ppm of the henna extract.

3. RESULTS AND DISCUSSION

3.1. Mass Loss (ML) Tests

The ML-time diagrams of C-steel coins in 1.0 M HCl, the presence and absence various concentrations of Henna extract, was measured after three hours of dipping at 25ºC have displayed in Fig. 2. % IE is recorded in Table 1. The presence of henna lowers the CR of C-steel in acid. The Figure shows that the henna actually inhibits the HCl corrosion of C-steel to a significant degree [16].

Table 1. ML parameters for dissolution of C-steel in HCl 1.0 M at various temperatures and at different concentrations of Henna extract

Temperature	Conc., ppm	ML, mg/cm²	Ɵ	% IE
25 ºC	Blank	6.50	--	--
	50	4.20	0.354	35.4
	100	3.70	0.500	50.0
	150	3.21	0.506	50.6
	200	3.09	0.525	52.5
	250	2.80	0.569	56.9
	300	1.20	0.815	81.5
35 ºC	Blank	11.16	---	---
	50	7.50	0.328	32.3
	100	7.20	0.355	35.5
	150	6.50	0.418	41.8
	200	5.90	0.471	47.1
	250	5.10	0.543	54.3
	300	3.18	0.715	71.5
45 ºC	Blank	28.45	--	--
	50	19.50	0.315	31.5
	100	18.60	0.346	34.6
	150	16.70	0.413	41.3
	200	15.10	0.469	46.9
	250	13.20	0.534	53.4
	300	8.89	0.688	68.8
55 ºC	Blank	57.34	--	--
	50	39.50	0.311	31.1
	100	37.90	0.339	33.9
	150	34.10	0.405	40.5
	200	31.30	0.454	45.4
	250	26.90	0.531	53.1
	300	20.3	0.641	64.1

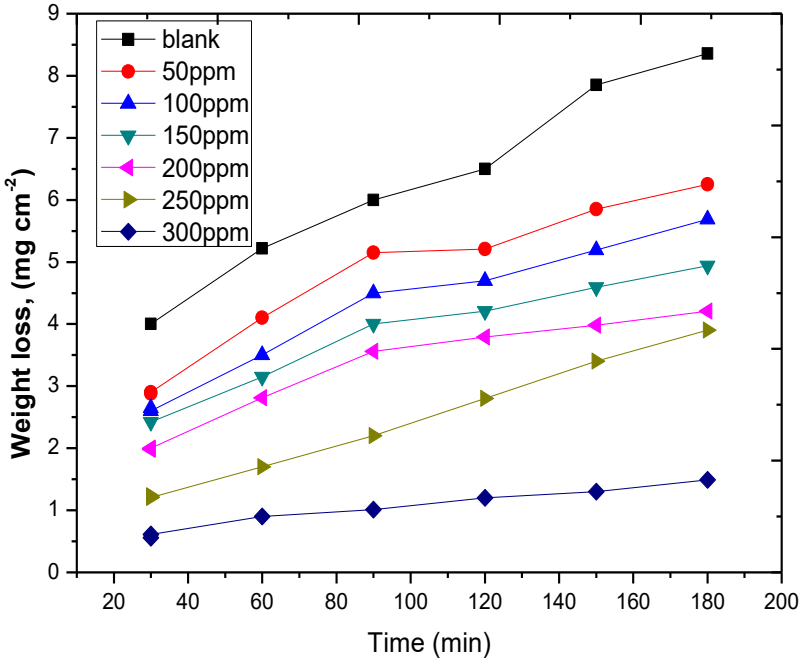


Figure 2. ML-time plots for C- steel corrosion in HCl 1M with and without different concentrations of Henna extract at 298 K

3.2. Adsorption Parameters

Several isotherms were utilized on the results, but the best one was Langmuir isotherm that identified as the next:

$C/\theta=1/K_{ads}+ C$ (5)

where K_{ads} is adsorption constant and C is extracted concentration Figure 3 display diagrams of (C/θ) against C at various temperatures. (ΔG^{o}_{ads}) were measured from Eq. (6):

$\log K_{ads} = - \log (55.5) - \Delta G^{\circ}_{ads} / (2.303RT)$ (6)

Figure (4) shows the plot of ΔG°_{ads} vs. T, as in Figure 4 the intercept gives the ΔH°_{ads} and the slope gives ΔS°_{ads} according to Eq. (7):

$\Delta G^{\circ}_{ads} = \Delta H^{\circ}_{ads} - T\Delta S^{\circ}_{ads}$ (7)

The obtained adsorption data were recorded in Table 2. Adsorption is spontaneous due to ΔG^{o}_{ads} has negative sign. Usually, values of ΔG°_{ads} up to -20 kJ mol^{-1} (physical adsorption) while those more negative than -40 kJ mol^{-1} (chemisorption) [17, 18]. The ΔG°_{ads} data were almost -32.9 kJ/mol, demonstrating the presence of physical and chemical adsorption (mixed adsorption). Negative ΔH^{o}_{ads} specified exothermic adsorption process [19]. ΔS^{o}_{ads} data were negative as accompanied by an exothermic adsorption procedure [20].

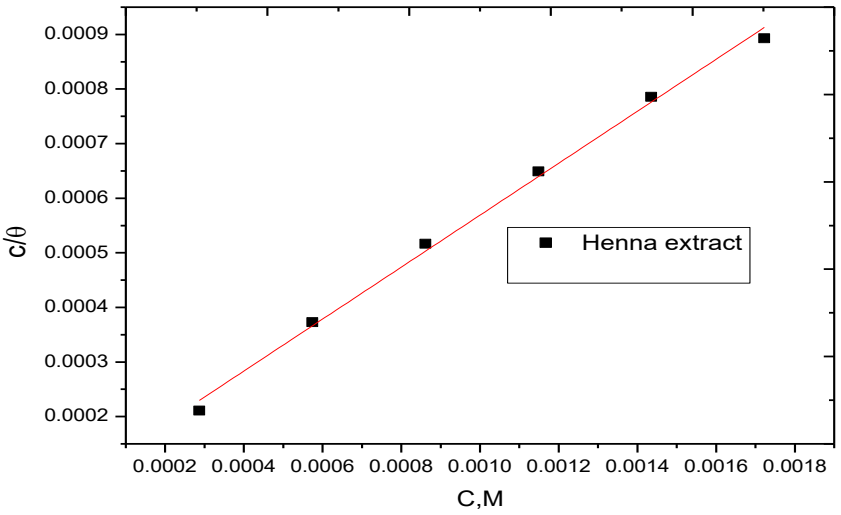


Figure 3. Langmuir plots for C-steel in HCl at 298 K

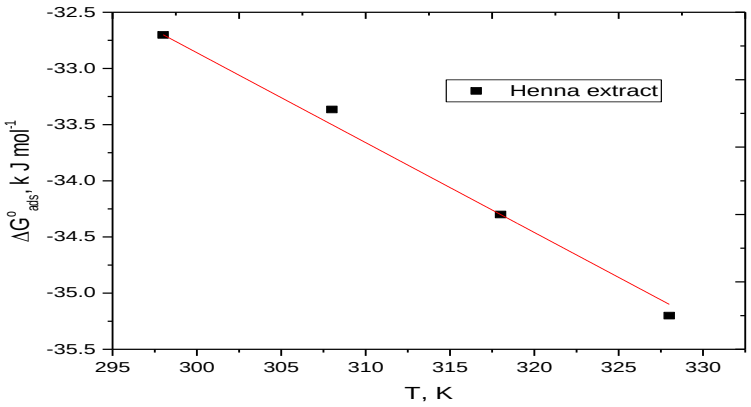


Figure 4. Plots of ΔG°_{ads} vs T for Henna extract at different temperatures

Table 2. Adsorption parameters of C-steel in corrosive solution for Henna extract

Inhibitor	Temp., K	$-\Delta S^{\circ}_{ads}$ J $mol^{-1}K^{-1}$	$-\Delta H^{\circ}_{ads}$ kJ mol^{-1}	$-\Delta G^{\circ}_{ads}$ kJ mol^{-1}	$K_{ads} \times 10^{+3}$ M^{-1}
Henna	298	30.7	42.5	33.0	10.57

3.3. Effect of Temperature

The influence of temperature on the rate of corrosion of C-steel in corrosive solution and in the presence of various concentrations (50-300 ppm) was examined in the range of temperature 25– 55ºC utilizing ML test. It was established that after 3 h dipping period, the IE lower with the rise in temperature (Table 1).

Activation energies for corrosion process (E^*_a) were obtained from the Arrhenius relation as Eq. (8):

$$\log k_{corr} = \log A - (E^*a / 2.303R) (1/T) \quad (8)$$

where A is constant, and T is Kelvin temperature [21-24]. Plots of log k_{corr} and 1000/T were illustrated in Figure 5. (ΔH^*) and (ΔS^*) were determined by plotting log k_{corr}/T against 1/T (Figure 6), according to the next relation:

$$\log k_{corr}/T = \log (R/ Nh + \Delta S^*/ 2.303R) + (-\Delta H^*/ 2.303R) (1/ T) \quad (9)$$

where h is constant and N is the number of Avogadro. Increasing of E^*_a and ΔH^* with Henna extract was because energy barrier that formed in the presence of Henna extract. ΔH^* values were found to have positive signs, indicating anodic dissolution reaction of C-steel. Negative ΔS^* indicated that from reactants to the activated complex, the disorder lowered [25].

Figure 5 shows a plot of (log k_{corr}) against (1/T) in the case of Henna extract in HCl. A straight line is obtained with a slope equals to (ΔH^* /2.303R) and the intercept is [log (R/Nh + ΔS^*/2.303R)] are measured (Table 3).

Table 3. C-steel kinetic parameters in the corrosive solution and in the presence of 150 ppm of Henna extract

Inhibitors	Conc. ppm	Activation parameters		
		E_a^*, kJ mol^{-1}	ΔH^*, kJ mol^{-1}	$-\Delta S^*$, J $mol^{-1}K^{-1}$
Blank	0.0	42.0	21.0	155.3
Extract	150	46.3	45.9	120.8

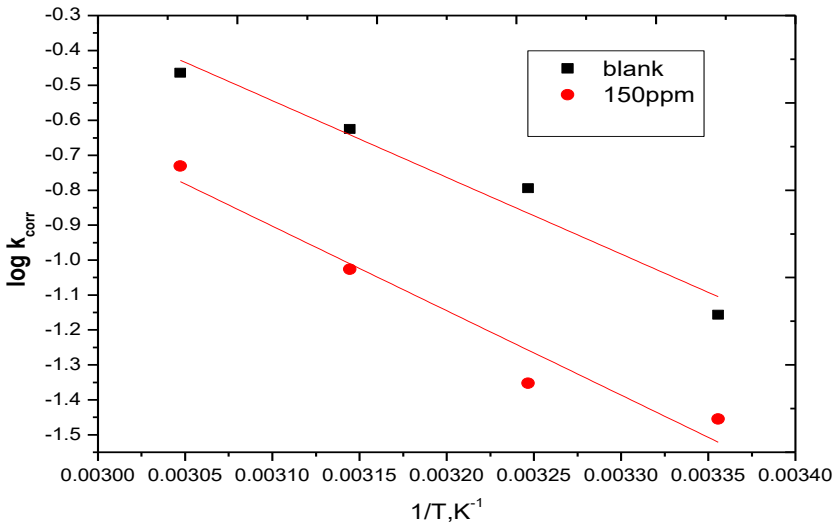


Figure 5. (log k_{corr}) vs. (1/T) for dissolution of C-steel in the corrosive solution and in the presence of 150 ppm Henna extract

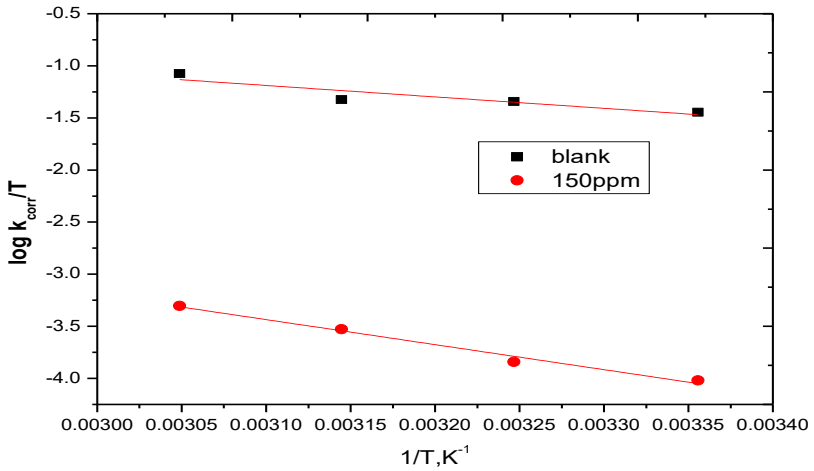


Figure 6. (log k_{corr}/T) vs. (1/T) for dissolution of C-steel in the corrosive solution and in the presence of 150 ppm Henna extract

3.4. Potentiodynamic Polarization (PP) Tests

Potentiodynamic Polarization diagrams for C-steel in unprotected and protected acidic solutions inclosing altered concentration of Henna extract are displayed in Fig.7. The parameters gotten from PP diagrams together with % IE are recorded in Table 4. The % IE increases as the rise the concentration of henna inhibitor. The small change in the β_a and, β_c (Tafel slopes) and in E_{corr} with raising the henna concentration indicates that this extract behaves as a mixed type [26]. The results data from PP (Table 4) show a decrease in i_{corr} with the presence of Henna extract and an increase in % IE with increasing the concentration. The parallel Tafel lines designate that there is no change in the mechanism of dissolution of C-steel.

Table 4. Potentiodynamic polarization parameters of C-steel with and without various of Henna extract at 298 K

Comp.	Conc *ppm*	-E_{corr}, *mV vs.SCE*	i_{corr} *mA cm^{-2}*	β_c *mV dec^{-1}*	β_a *mV dec^{-1}*	θ	% IE	k_{corr} *mpy*
Blank	0.0	449	959.0	236	160	---	---	252.9
Henna	50	459	235.0	168	95	0.755	75.5	107.5
	100	471	221.3	170	98	0.770	77.0	101.1
	150	474	201.4	161	114	0.790	79.0	89.7
	200	482	189.7	164	109	0.803	80.3	75.9
	250	486	173.1	163	114	0.820	82.0	70.3
	300	491	163.6	163	117	0.831	83.1	66.9

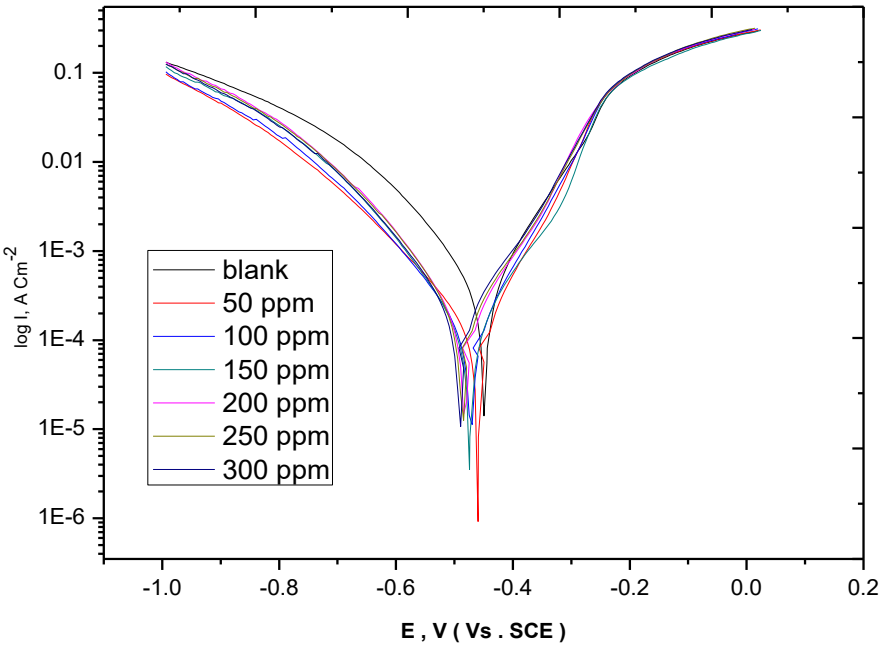


Figure 7. PP curves of C-steel in HCl with and without different concentrations of Henna extract at 298 K

3.5 Electrochemical Impedance Spectroscopy (EIS) Tests

The corrosion of C-steel in 1.0 M HCl in the presence and absence of various concentrations of Henna extract was examined by EIS test at 298 K. Impedance parameters are resulting from the Nyquist diagrams (Fig. 8) and are a record in Table (5). Nyquist plots designated that the presence of henna extract increased the semicircle diameter, which leads to the rising resistance of dissolution C-steel. It is detected that the data of R_{ct} improve with improving the concentration of the Henna extract and this in turn indications to a lower in the rate of C-steel dissolution in corrosive solution. The circuit shown in Figure 9 was used to fit EIS data [27-28]. The C_{dl} values lower with improving the concentration of Henna extract. C_{dl} was determined according to equation 9:

$$C_{dl} == [1/2\pi f_{max} R_{ct}] \quad (9)$$

where f is frequency maximum,

Table 5. EIS data of C-steel with and without various concentrations of Henna extract at 298 K

Comp.	Conc., ppm	C_{dl}, µF cm^{-2}	R_{ct}, Ω cm^2	Θ	%PE_{EIS}
Blank	0.0	132.4	28.5	---	---
Henna	50	113.2	62.9	0.547	54.7
	100	94.4	81.5	0.651	65.1
	150	80.2	90.2	0.684	68.4
	200	72.9	98.3	0.710	71.0
	250	68.4	107.4	0.734	73.4
	300	61.2	138.8	0.795	79.5

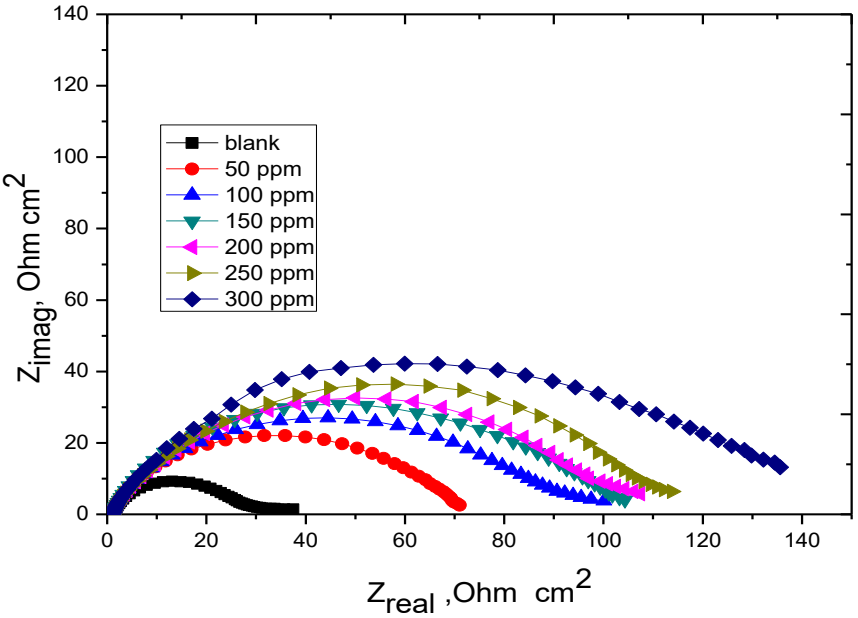


Figure 8. Nyquist diagrams of C-steel with and without various concentrations of Henna extract at 298K

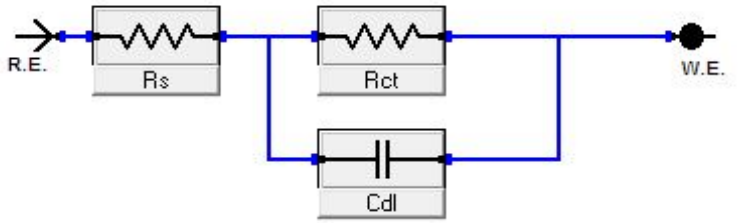


Figure 9. Circuit utilized to fitting EIS data

3.6. Electrochemical Frequency Modulation (EFM) Tests

Table 6. EFM parameters for C-steel with and without various concentrations of Henna extract at 298K

Conc., ppm	i_{corr} , μAcm^{-2}	β_c , $mVdec^{-1}$	β_a , $mVdec^{-1}$	CF-2	CF-3	θ	%IE	CR , mmy^{-1}
Blank	628.1	126	99	1.940	2.978	----	----	287.1
50	361.5	145	96	2.026	3.153	0.425	42.5	250.2
100	320.9	146	106	2.035	3.210	0.489	48.9	220.9
150	291.3	128	94	2.001	3.016	0.536	53.6	191.3
200	209.3	143	121	2.074	3.080	0.667	66.7	171.2
250	160.9	149	112	1.982	2.981	0.744	74.4	150.6
300	120.9	152	131	1.894	2.991	0.808	80.8	120.4

The EFM is a safety dissolution test that can directly obtain data of the corrosion current devoid of the preceding information of Tafel constants. Figure (10-11) demonstration EFM spectra for steel in hydrochloric acid and in presence of 150 ppm Henna, correspondingly. EFM data results (i_{corr}, β_c, β_a, causality factors (CF-2 & CF-3), k_{corr}, θ and %IE) were record in Table 6. It was clear that as Henna extract concentration, rise i_{corr} decreases and %IE rises. CF-2&CF-3 specified that the outcome data were of excellent quality. The outcome data displayed good agreement of %PE gotten from the PP, EIS and MR tests.

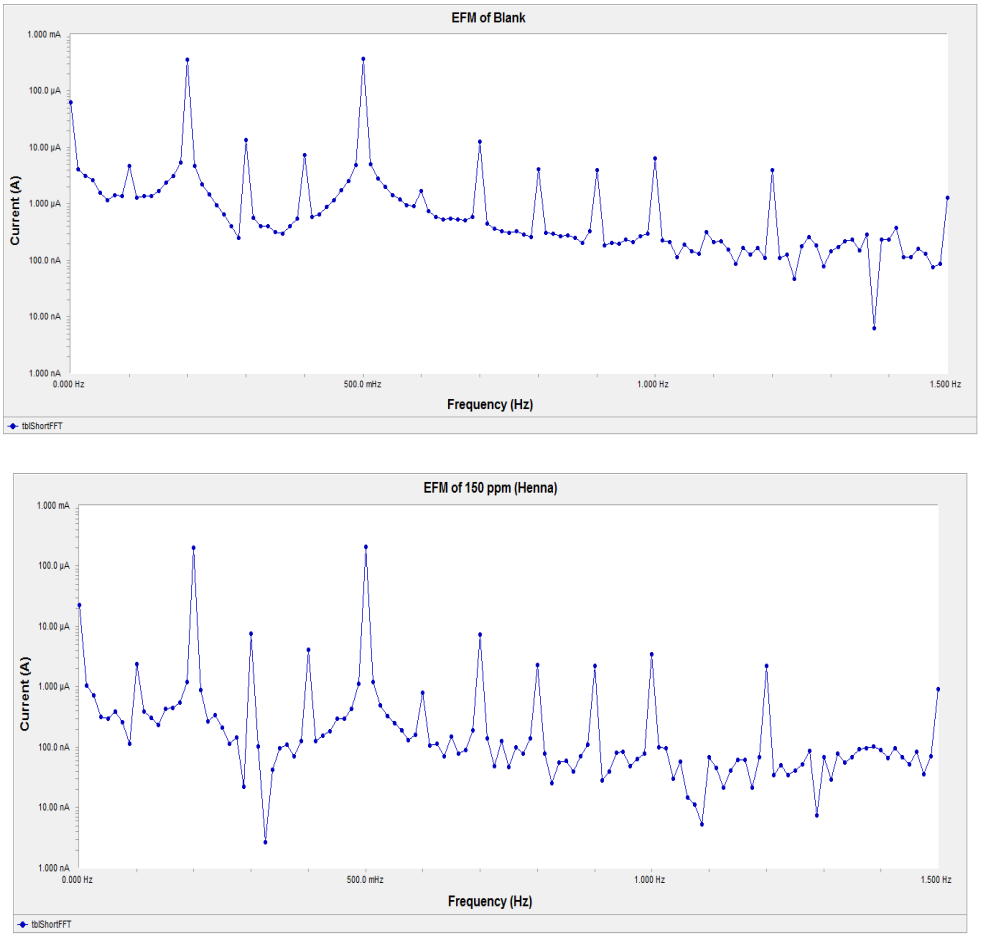


Figure 10. EFM diagrams for C-steel in HCl solution (a) and in the presence of 150 ppm Henna (b).

3.7. Energy Dispersion Spectroscopy (EDX) Studies

The EDX spectra were utilized to measure the presence of the element on the C-steel surface after three days of contact to the unprotected and protected 1M HCl. Fig.11 portrays the EDX study of C-steel in a corrosive medium only and in the presence of 300 ppm of extract. A comparable elemental distribution is presented in Table (7). The data indicated that there are C&O atoms on the free C-steel surface representative presence of Fe_2O_3 layer on C-steel surface, while in addition of henna extract, additional lines appeared due to adsorption Henna extract on C-steel.

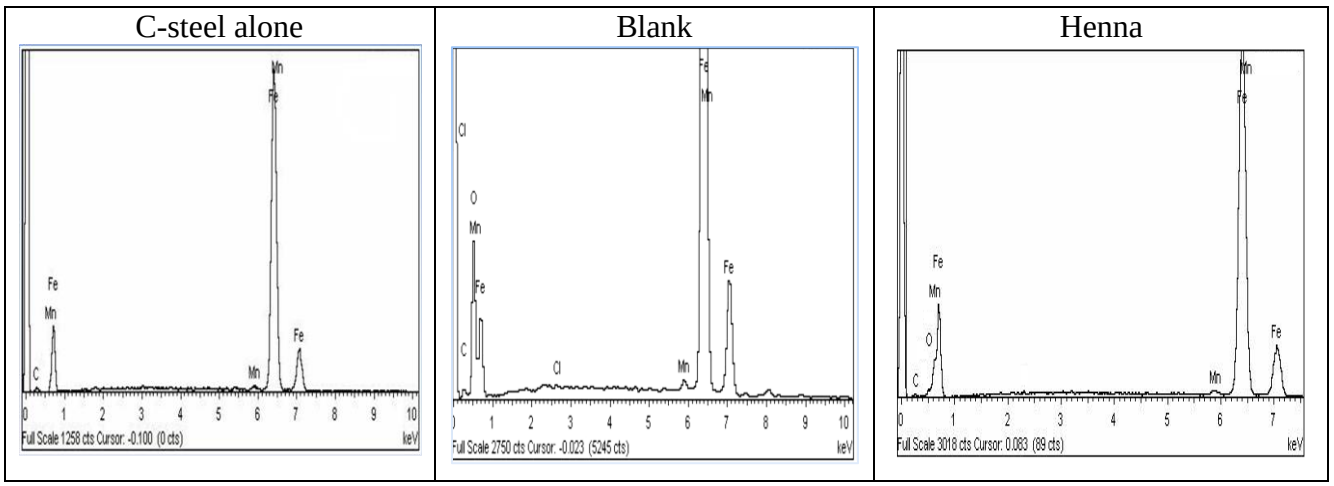


Figure 11. EDX plots for C-steel dissolution with and without 300 ppm Henna for immersion three days

Table 7. C-steel mass % of after three days of immersed in HCl with and without 300 ppm Henna extract

(Mass %)	Fe	Mn	C	O	N	Cl
C-steel	96.78	0.61	4.87	--	--	--
C-steel + HCl	55.83	0.31	2.12	39.24	--	0.32
C-steel+HCl+extract	61.23	0.43	19.85	10.18	--	-

3.8. SEM Tests

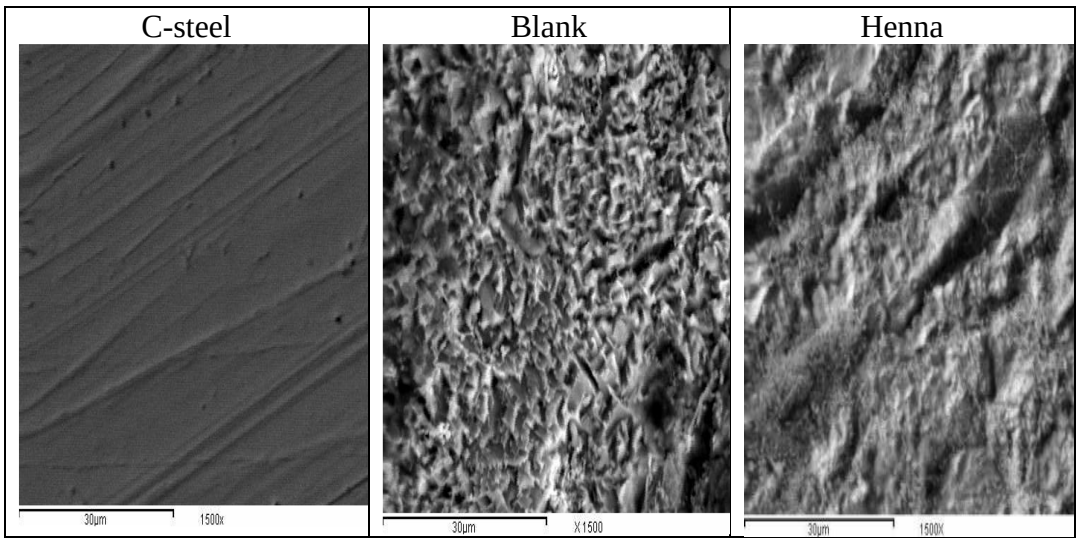


Figure 11. SEM micrographs for C-steel with and without 300 ppm Henna

Figure (12) signifies the micrograph obtained from samples of C-steel after contact with a corrosive solution for dipping three days. It is observed that C-steel suffers from hard attack corrosion in HCl. While the presence of henna extract reduces the damage of C-steel surface and makes the surface

smoother. Because formation of a passive layer through adsorbed henna on the CS surface that blocks the active sites and lower corrosion [31-32].

3.9 Mechanism of Inhibitor of C-steel

The result designates that extract (henna leaves) achieves as an excellent inhibitor for the corrosion of C-steel in corrosive solution. It was described that extract (henna leaves) includes soluble matter Lawson. Lawson compound is a ligand that can chelate with various metal cations creating complex. Fig. 11 shows the rearrangement of Lawson molecule due to the presence of the hydroxyl group which contains a pair of electrons, which is delocalized in a hydrochloric acid [33]. This rearrangement gives stabilized metal-complexes. The great IE of the henna extract in HCl for C-steel can be accepted by the formation complexes among Lawsonia molecules and Fe^{2+} cations present in the corrosive solution due to the dissolution of C-steel. Then this complex can be adsorbed on metal surfaces and cover larger areas. To have a comparative analysis of the present work to that with some other plant extracts, Table 5 is given below. In general, the %IE of the extract used in the current work is considerably higher for the C-steel.

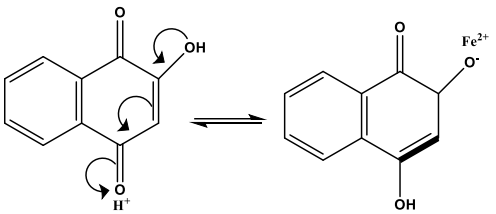


Figure 11. Rearrangement of Lawsonia

Table 5. Comparison of inhibition efficiencies of C-steel with other plant material extracts in HCl media

Inhibitor (Extract)	IE %	References
Henna leaves	83.1	Our results
Sida acuta	80.0	[34]
Raphanus sativus	80.2	[35]
Ginger affinal	63.4	[36]
Ginkgo biloba	82.0	[37]
Dendrocalmus sinicus	79.2	[38]
Cassia italica	84.9	[39]
Anacyclus pyrethrum root	79.0	[40]
Aerial part of Salvia aucheri mesatlantica	70.1	[41]
Loquat leaves	74.9	[42]

4. CONCLUSIONS

The Henna extract shows the corrosion inhibitor for C-steel in hydrochloric acid solution, where the PE ratio improved by the rise of henna concentration. The PE is decreasing with the rise of temperature resulting from the destruction of the adsorbed henna molecules present on the C-steel surface. The presence of Henna on the surface follows the Langmuir equation. Tafel curves showed that henna extract is mixed-kind inhibitors. (C_{dl}) reduced by the rise of the henna concentration. While (R_{ct}) rise. The adsorbed passive layer on the C-steel surface was proved by SEM and EDX analysis.

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