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The Corrosion Inhibition and Adsorption Properties of Eco Friendly Green Inhibitor – A Comparative Study

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Abstract : The corrosion inhibition and adsorption behaviour of aqueous extract of plants like Justicia micrantha (JM), Hibiscus callyphyllus (HC), Morinda tinctoria (MT), Poinciana Elata (PE), ErythrinaIndica (EI), Sphoeranthus mirtes (SM) on mild steel surface in 1N HCl solution were investigated by mass loss with different time of contact at various temperature and evaluated by electrochemical impedance and Tafel studies. Polarization method indicates that the plant extract was a mixed type inhibitor with predominately control of anodic reaction. The nature of Protective film formed on the MS surface has been confirmed by SEM analysis.
Keywords : Mild steel; EIS; SEM; Polarization; Acid Corrosion.

1. Introduction

Mild steel is world widely used in many application in industries like metallurgical process, chemical cleaning, fertiliser, desalination plants, nut bolts, screw industry, storage tank, petroleum refineries, pharmaceutical industry, thermal power point, construction material, and gas industry, sugar, paper, textile, boilers, automobiles, steam boilers, engineering purpose, mechanical purpose, chain, hinges, knives, magnets, military equipment, armour, vehicles (ships and cars)and pickling process etc., due to their stability, high strength, weld ability and good corrosion resistance[1-5]. Corrosion is only nature's method, inevitable and serious problem, because it definitely contributes to the depletion of our natural resource and non - stop. That problem faced by almost all alloys and industries, can be considered as one of the worst environmental calamite of our time. Most metals are commonly unstable in the atmosphere, both dangerous and costly. Every year each country spends billions of dollars for the replacement of corroded component and machinery structure in direct and indirect cost wise. Literature survey reveals that and have been reported for synthetic Organic[6-10], polymer[11-17], hetero cyclic compounds[18-25]gum, bakery, industry waste, plants extract[26-35], and various parts like leaf, flowers, seeds, fruits[36-39] as corrosion inhibitor for (mild steel, Al, carbon steel, Zn, Cu and Alloys) in various medium[40-45]. The recent researcher reported that the plant extract that can be used as an environmental or green corrosion inhibitor for protecting metal against corrosion. The plant material of natural product origin is inexpensive, environmental friendly, cheap, easily available, renewable source, ecologically acceptable, easy handling and simple procedure[46-52].The use of aqueous extract of (Justicia micrantha (JM), Hibiscus callyphyllus (HC), Morinda tinctoria (MT), Poinciana Elata (PE), Erythrina indica (EI), Sphoeranthus mirtes (SM)) plant as a green inhibitor has not been reported elsewhere. In continuation of our research work, the present investigation deals with the inhibitive and adsorption properties of aqueous extract of (JM, HC,MT, PE, EI, SM) leaves for the corrosion of mild steel in HCl medium of different time and different temperature.

Material and methods

Specimen preparation

MS coupons having composition in weight (%)Mn- 0.169, Mb- 0.016, C- 0.030, Cr- 0.029, P- 0.031, S - 0.029, Ni- 0.030, Cu- 0.017, Si- 0.015 , and reminder Fe with a hole near upper edge (3mm diameter) have been used for corrosion inhibitor study.

Inhibitor preparation

Freshly collect green leaves were first cleaned, dried and ground fine powder. The extracts were prepared by refluxing 25g of each powder and add 250ml of distilled water and heated for 3 hours and kept overnight. This prepared solution was used to study the corrosion inhibition properties.

Mass loss method

Mass loss measurement is a classical way to determine the corrosion inhibition and corrosion rate of mild steel. MS Coupons were completely immersed vertically in 150 ml of the test solution with and without inhibitor for 24 hours. The experiment was carried out at various immersion period (1, 3, 5, 7, 12h) and corrosion inhibition studies were also carried out at various temperature ranges at(278-323 K).

$$IE\ \% = \frac{W_o - W_i}{W_o} x100 \qquad (1)$$

W_0 and $W_i$are the weight loss with and without of the inhibitor.

2.4. Electrochemical methods

The polarization measurements were carried out on an electrochemical cell with a three electrode cell set up was used. Mild steel ($1cm^2$) was used as a working electrode; Pt electrode was used as counter electrode, and a saturated calomel electrode as reference electrode. The electrode setup was fully and separately immersed in the (acid) solution for approximately 30 minutes to attain open circuit potential (OCP). Anodic and cathodic current – potential curves were recorded by polarization from - 0.5 mV to +0.5 mV at a scan rate of 1 mVs^{-1}.

$$IE\ \% = \frac{I_{Corr} - I_{*Corr}}{I_{Corr}} x\, 100 \qquad (2)$$

I_{corr} and I_{*corr} are corrosion current in the without and with inhibitors.

Electrochemical impedance

Polarization studies were done immediately after EIS studies on the conventional three electrode cell assembly without any further surface treatment. A plot of Z' versus Z" was made. The double layer capacitance (C_{dl}) was determined using formula:

$$C_{dl} = \frac{1}{2\pi} f_{max} R_{ct} \qquad (3)$$

R_{ct} is charge transfer resistance, and C_{dl} is double layer capacitance.

Result and Discussion

Mass Loss Method

Mass loss analysis is one of the basic, easy and frequently used methods of determining corrosion of metal in the given medium. Corrosion rate and inhibition efficiency of plant extract on mild steel at different immersion time and various temperature data are presented in Table 1 and 2.It is revealed from the table that the presences of phytochemical constituent in the plant extract are found to be bigger molecules to cover a larger surface area on adsorption. Most of the corrosion inhibitor act by adsorption of their molecules on the metal surface. However, in temperature variation the inhibition efficiency decreases with increase of temperature which indicates the inhibitor film formed on the metal surface is less protective in nature at higher temperature because of desorption of inhibitor molecules from the metal surface[53-57]. These results evidently showed that all the six plants extract exhibit the most potential anticorrosive properties.

Table 1.Percentage of inhibition efficiency (IE %) and corrosion rate (CR) at different concentration of inhibitor in 1N HCl medium as a various immersion time.

Plant name	Inhibitor (v/v)	Inhibition efficiency (%) 1h	3h	5h	7h	9h	12h	24 h
Justicia micrantha	Blank	-	-	-	-	-	-	-
	5	58.01	61.33	56.16	34.00	64.20	42.26	65.31
	10	68.19	70.25	68.74	56.76	70.12	58.72	78.03
	15	77.10	79.69	76.06	65.96	72.49	66.95	86.82
	20	80.20	88.64	83.57	80.57	81.38	84.90	90.83
Hibiscus callyphyllus	5	60.31	54.90	70.13	76.16	65.78	70.21	50.90
	10	65.78	66.59	72.08	77.12	66.78	74.78	56.11
	15	78.25	78.90	86.00	84.67	89.62	85.23	69.78
	20	94.33	91.23	94.15	93.99	96.18	99.91	79.59
Morinda tinctoria	5	62.11	68.10	66.14	64.51	73.90	88.20	75.33
	10	69.65	79.66	75.83	72.68	77.48	89.93	95.90
	15	77.54	86.12	89.80	82.99	88.79	91.18	86.28
	20	83.11	95.49	98.99	95.97	95.34	97.35	89.42
Poinciana elata	5	77.70	78.33	86.36	86.56	88.27	87.61	98.57
	10	85.90	87.44	88.83	89.98	90.96	91.56	78.57
	15	86.48	88.74	89.59	90.98	95.18	93.65	70.23
	20	94.62	96.34	94.85	98.09	97.54	99.60	69.51
Erythrina indica	5	45.12	55.21	66.21	60.45	54.42	69.21	79.64
	10	54.33	56.78	78.90	68.90	67.54	78.21	84.16
	15	68.45	79.89	81.45	77.67	79.09	85.90	89.04
	20	79.34	87.23	90.23	84.06	89.54	94.56	96.45
Sphoeranthus mirtes	5	34.23	56.89	54.78	57.09	67.90	78.33	76.28
	10	45.56	66.09	56.67	64.23	78.45	82.90	89.42
	15	65.98	78.45	89.45	78.89	87.43	88.78	98.55
	20	88.90	87.34	90.45	89.43	94.32	96.71	94.57

Table 2.ThePercentage inhibition efficiency of Selected six Plants leaves extract at various temperatures

Plant name	the extract (v/v)	IE (%) 303K	313K	323K
Justicia micrantha	Blank	-	-	-
	5	24.10	29.20	56.76
	10	45.60	37.27	62.24
	15	56.18	43.55	79.66
	20	66.67	72.34	83.05
Hibiscus callyphyllus	5	46.26	45.30	54.25
	10	61.11	59.20	58.34
	15	76.38	65.07	64.26
	20	89.12	87.45	85.79
Morinda tinctoria	5	34.15	59.85	51.67
	10	38.69	63.17	69.49
	15	52.55	67.40	80.05
	20	78.76	81.58	85.45
Poinciana	5	27.50	32.55	46.67

elata	10	40.52	49.89	60.77
	15	57.91	78.87	68.56
	20	79.60	85.02	89.03
Erythrina indica	5	14.34	34.89	29.56
	10	39.23	56.78	45.89
	15	53.12	79.40	58.92
	20	78.60	91.09	72.54
Sphoeranthus mirtes	5	20.14	45.67	56.92
	10	40.45	59.90	67.89
	15	56.78	78.34	74.27
	20	69.09	89.23	83.99

Table 3.Electrochemical parameters from polarization measurement and calculated values of inhibition efficiency.

Plant name	Conc. (v/v)	E_{corr}/ (mV/ SCE)	I_{corr}/ (mA/cm^2)	b_c (mV/dec.)	b_a (mV/dec.)	IE (%)
Justicia micrantha	Blank	-0.4712	$6.946x10^{-3}$	87	128	*
	5	-0.4726	$1.479x10^{-3}$	72	138	77.53
	10	-0.4729	$1.370x10^{-3}$	72	138	76.58
	15	-0.4748	$1.114x10^{-3}$	66	135	83.58
	20	-0.4556	$1.620x10^{-4}$	65	90	76.08
Hibiscus callyphyllus	Blank	-0.4712	$6.946x10^{-3}$	87	128	*
	5	-0.4453	$1.804x10^{-3}$	61	150	74.06
	10	-0.4466	$1.309x10^{-3}$	64	143	79.93
	15	-0.4477	$1.450x10^{-3}$	62	154	81.26
	20	-0.4522	$8.359x10^{-5}$	64	134	88.09
Morinda tinctoria	Blank	-0.4712	$6.946x10^{-3}$	87	128	*
	5	-0.4684	$5.630x10^{-5}$	160	64	18.87
	10	-0.4535	$2.163x10^{-3}$	68	147	68.87
	15	-0.4555	$1.129x10^{-3}$	68	134	83.86
	20	-0.4581	$9.390x10^{-5}$	69	129	86.58
Poinciana elata	Blank	-0.5046	$6.946x10^{-3}$	87	128	*
	5	-0.3450	$5.425x10^{-3}$	127	87	86.31
	10	-0.3676	$7.301x10^{-7}$	116	97	89.48
	15	-0.3653	$1.065x10^{-6}$	126	89	84.87
	20	-0.3787	$9.677x10^{-7}$	120	95	86.16
Erythrina indica	Blank	-0.5046	$6.946x10^{-3}$	87	128	*
	5	-0.1118	$2.860 x10^{-6}$	38	202	19.52
	10	-0.1037	$3.501 x10^{-6}$	42	281	21.75
	15	-0.0989	$5.43 x10^{-6}$	38	252	49.56
	20	-0.1138	$5.58 x10^{-6}$	41	261	58.78
Sphoeranths mirtes	Blank	-0.5046	$6.946x10^{-3}$	87	128	*
	5	-0.3432	$3.260 x10^{-7}$	101	111	41.46
	10	-0.3401	$3.140 x10^{-7}$	110	100	51.89
	15	-0.2784	$6.845 x10^{-7}$	113	89	53.02
	20	-0.2748	$6.485 x10^{-7}$	113	89	54.75

Table 4. Impedance parameter for mild steel in 1 N HCl acid solution in the absence and presence of varied concentration of inhibitor

Plant name	Concentraion (v/v)	R_{ct} (ohm cm^2)	C_{dl} ($\mu F/cm^2$)	IE (%)
Justicia micrantha	Blank	10	1.57 $x10^{-5}$	*
	5	33	$4.69x10^{-5}$	69.69
	10	45	$4.68x10^{-5}$	77.77
	15	66	$3.06x10^{-5}$	84.84
	20	72	$4.43x10^{-5}$	86.11
Hibiscus callyphyllus	Blank	10	1.57 $x10^{-5}$	*
	5	19	$4.90x10^{-6}$	47.36
	10	42	$1.35x10^{-5}$	76.19
	15	52	3.03 $x10^{-5}$	80.76
	20	64	$7.08x10^{-5}$	84.37
Morinda tinctoria	Blank	10	1.57 $x10^{-5}$	*
	5	26	$6.85x10^{-6}$	61.53
	10	45	$9.48x10^{-6}$	77.77
	15	58	$5.23x10^{-5}$	82.78
	20	67	$6.23x10^{-5}$	85.07
Poinciana elata	Blank	10	1.57 $x10^{-5}$	*
	5	34	$9.18x10^{-5}$	70.58
	10	39	$2.15x10^{-5}$	74.35
	15	67	$5.33x10^{-1}$	85.07
	20	72	$1.01x10^{-6}$	86.11
Erythrina indica	Blank	10	1.57 $x10^{-5}$	*
	5	39	2.50 $x10^{-4}$	74.35
	10	56	1.26 $x10^{-4}$	82.14
	15	72	7.41 $x10^{-5}$	86.11
	20	84	7.40 $x10^{-5}$	88.09
Sphoeranthu smirtes	Blank	10	1.57 $x10^{-5}$	*
	5	38	4.09 $x10^{-5}$	73.68
	10	49	1.63 $x10^{-4}$	79.59
	15	65	2.97 $x10^{-5}$	84.61
	20	71	1.19 $x10^{-4}$	85.91

Potentiodynamic polarization methods

Figure 1 represents the polarization curves for MS in 1NHCl solution containing different concentration of selected six plant extract and values derived from these investigations are given in table 4. It is observed from the table that the inhibition efficiency has occurred due to simple blocking of the active sites, thereby reducing available surface area of the corroding metals[58].

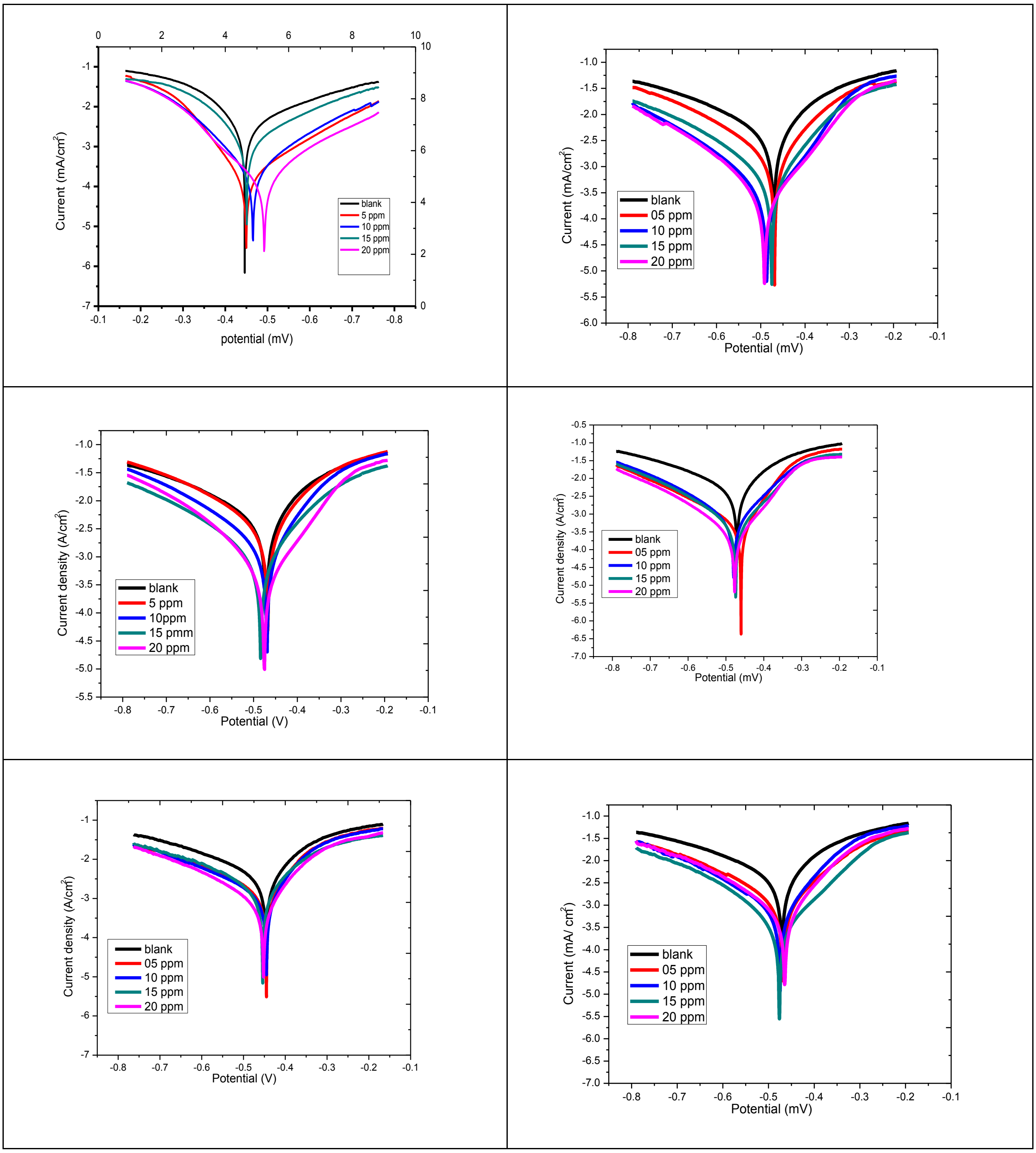


Fig. 1 Potentiodynamic polarization (Tafel) curves for mild steel in 1N HCl solution in the absence and presence of different concentration of *different extracts* of leaves.

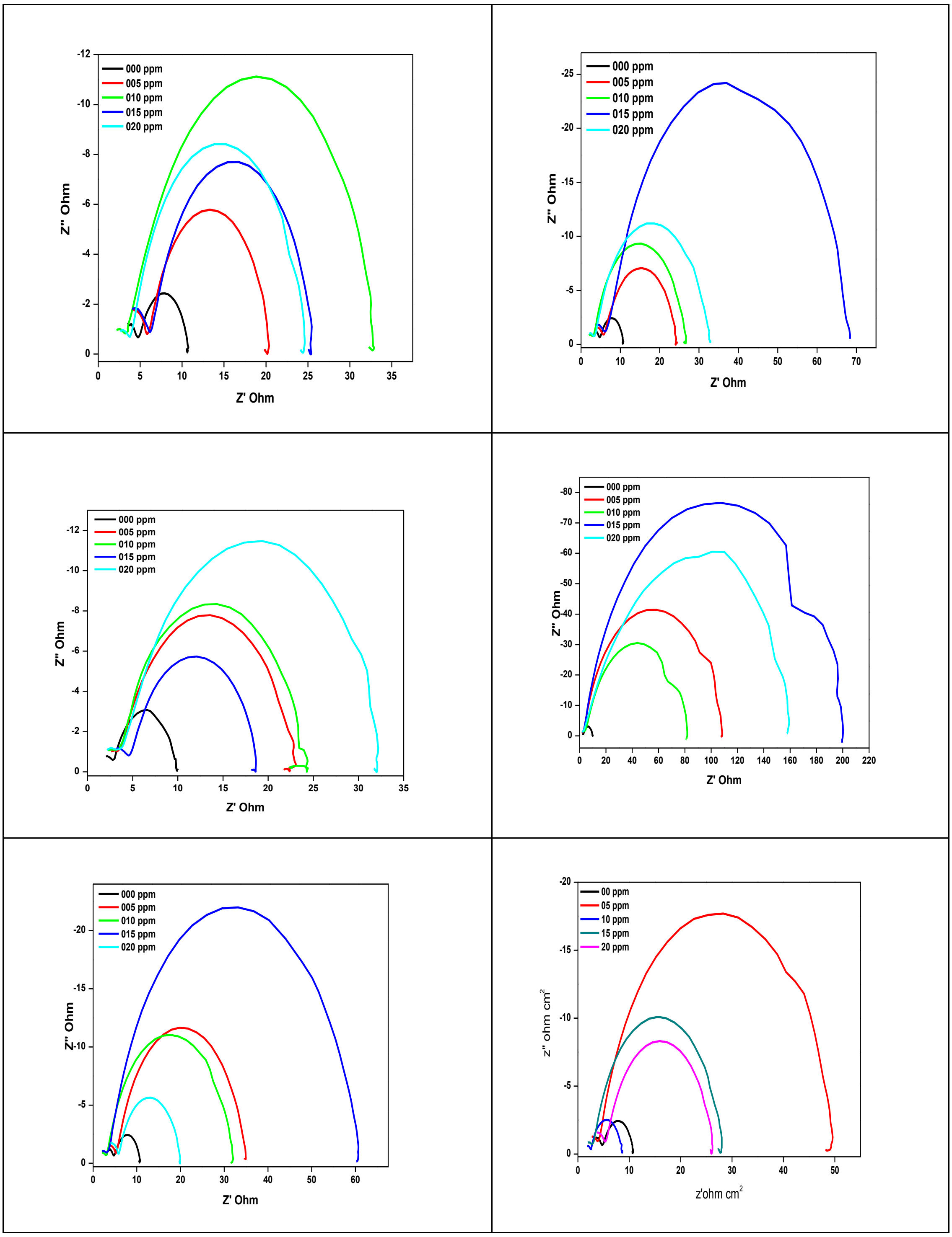


Fig. 2 Nyquist plots for mild steel in 1N HCl acid solution without and with presence of different concentration of *different* extract of leaves

Electrochemical impedance method

Nyquist plots of MS in uninhibited and inhibited test solution containing various concentrations of all the plants leaves extract are presented in figure 2 and EIS parameter are listed in table 5. The impedance data fit the Nyquist curves clearly indicates that the typical characteristic of suppressed semi - circular behaviour, but increasing semi - circular of diameter proved increasing protection against corrosion. All the electrochemical parameters clearly proposed that the addition of extract does not affect the mechanism of mild steel dissolution and corrosion control depends on the concentration of the inhibitor[59].

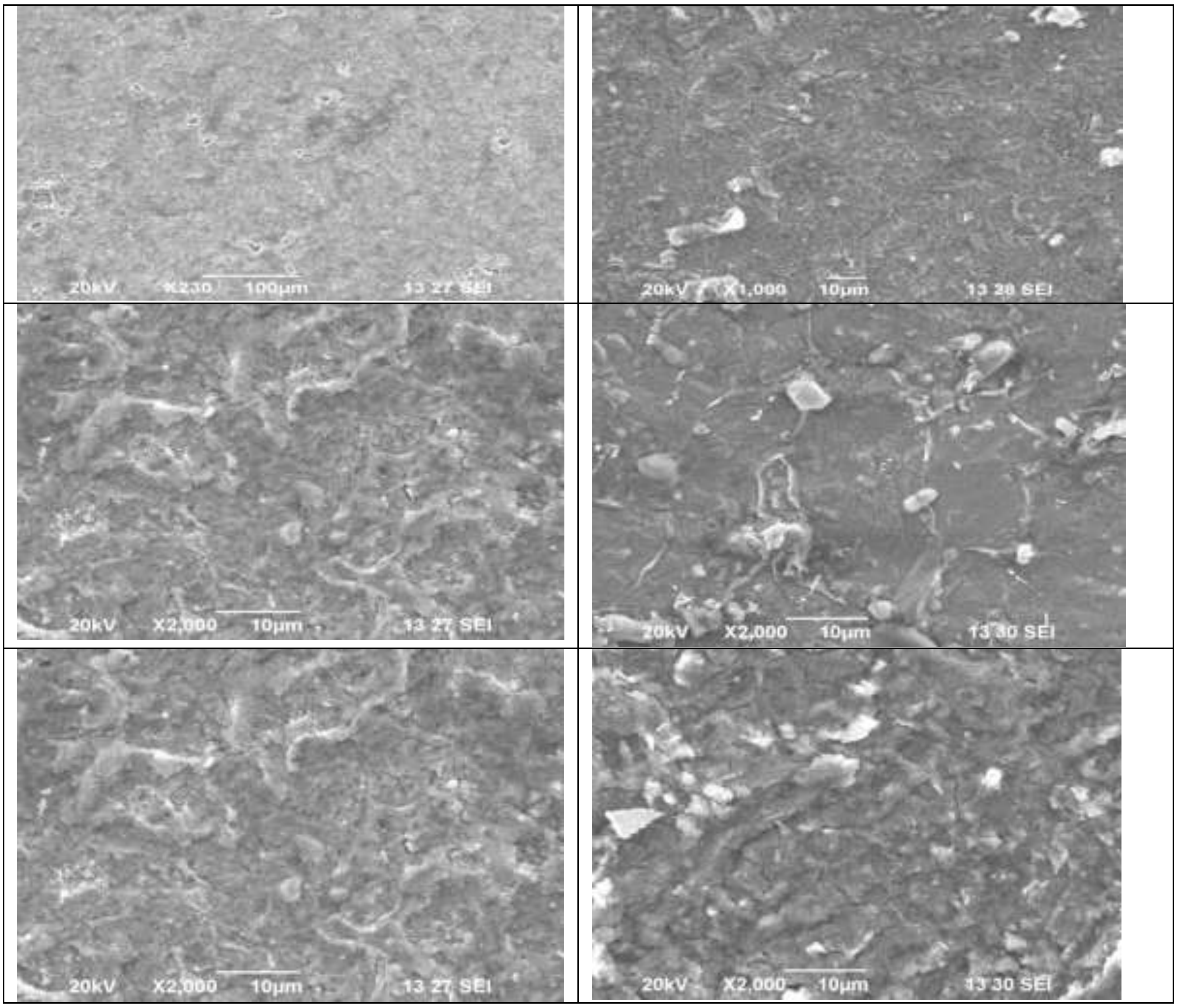
Table 5.Phytochemical screening test for extract of select six plants leaves extract.

Phytochemical test	JM	HC	PE	MT	SM	EI
Alkaloids	+	+	+	+	+	+
Carbohydrates	+	+	+	+	+	+
Proteins	+	+	+	+	+	-
Saponins	-	-	+	+	-	+
Thiols	+	-	_	+	-	-
Tannins	-	-	+	+	+	-
Flavanoids	-	+	+	-	-	+
Phenol	-	+	+	-	+	-
Glycosides	-	+	+	+	-	+

(+).. Presence (-)... Absence

Phytochemical screening test

In view of its phytochemical investigation, it was much cleared that the presence of phytochemical is responsible for the anticorrosion action. The screening test was carried out on a freshly prepared extract and various chemical constituent are presented. The result indicates that the natural product (alkaloids, tannings, steroids, terpinoids) absolutely inhibited the corrosion product on the metal surface.



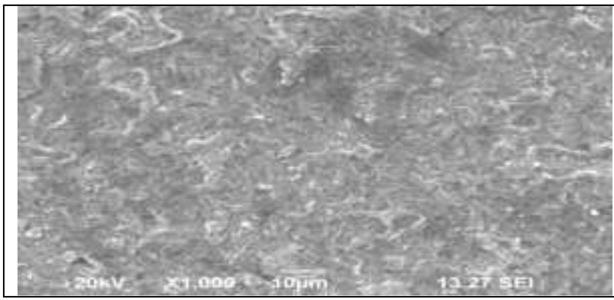


Fig. 3 SEM image of the surface of mild steel after immersion for 24 hours in 1N HCl solution absence (3A) and in the presence (3B- 3G) of optimum concentration of the different plant Leaves extracts.

Surface analysis

The observation of the mild steel specimen was carried out using scanning electron microscope. Figure 3 shows the SEM image of mild steel surface after immersed in 1NHCl in the absence and presence of allselectedsixplant extract for 24 hours. On closely observed, very strong corroded (pits and crack) and uneven (heavy damage) metal surface obtained when the metal was kept immersed in 1N HCl for absence of inhibitor. In presence of inhibitor the metal surface shows smoother lager with clearly different morphology (surface covered means no pits and cracks).

Adsorption isotherm

The information on the collaboration between inhibitor molecules (organic adsorbate) and mild steel surface can be provided by adsorption isotherm. In order to evaluate the adsorption process of selected six plant extract on the metal surface Temkin, Langmuir, Hasley, Freundlish, El-awardy, Parson, sFrumkin were made to find the best isotherm which describes in this studyThe straight line in figure 4 clearly indicated that the inhibitor obey Temkin adsorption isotherm. Organic compounds usually contains polar function with hetero atom such as N, S, O,P and have double or triple bond or aromatic ring. Good correlation between plant water soluble constituent and suggest physical adsorption mechanism was obtained.

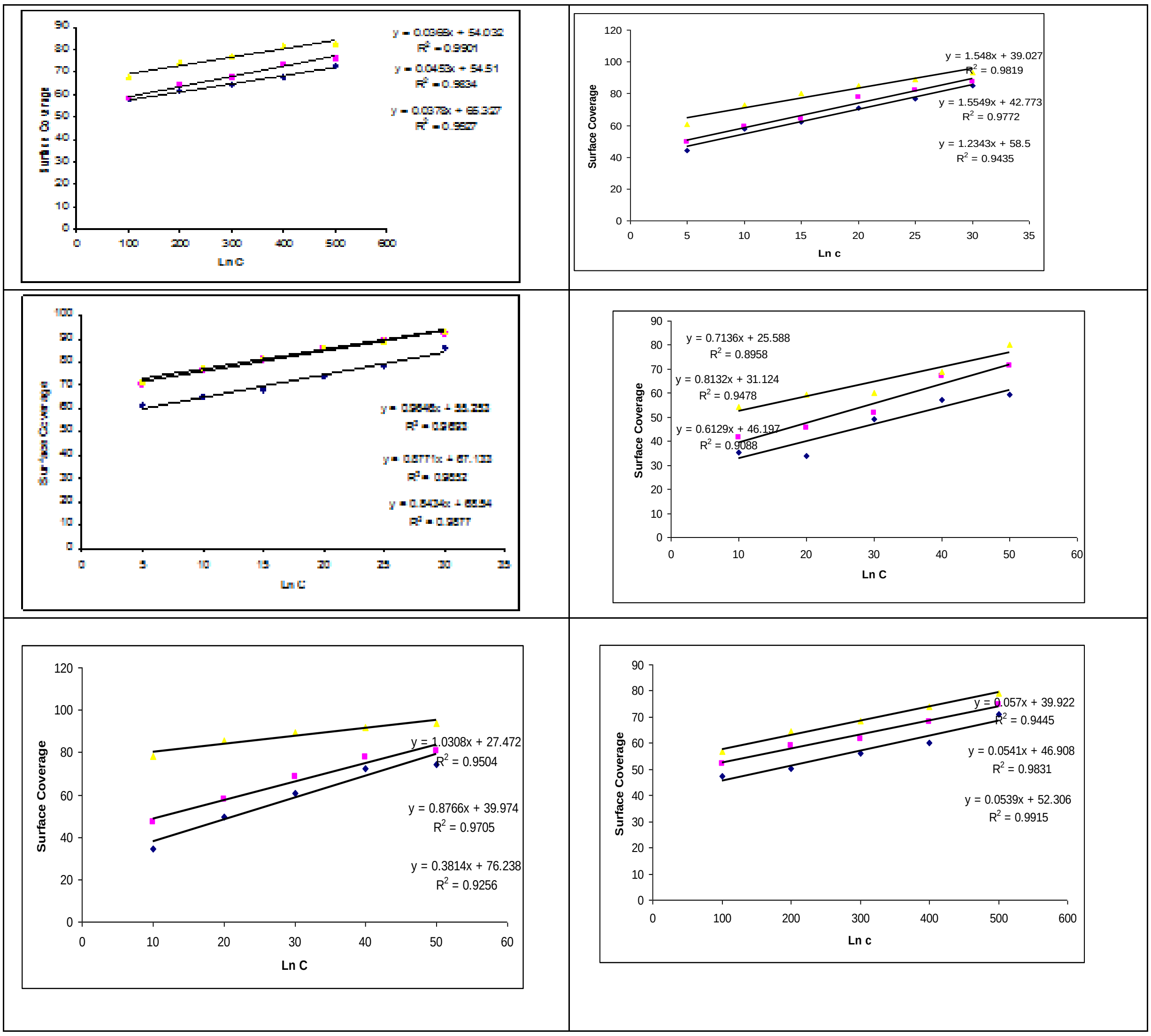


Fig. 4 Temkin adsorption isotherm plot for mild steel in 1N HCl containing different concentration of the extract.

Conclusion

The selected six leaves extract can serve as an excellent eco - friendly green inhibitor for the corrosion of mild steel in 1N Hydrochloric acid. The inhibition efficiency of the extract increases with increase in the concentration and the inhibition may due the formation of protective film on the mild steel surface.The inhibition efficiency values obtained from both chemical and electrochemical methods (Weight loss, polarization and impedance measurement) are in good agreement with each other. Potentiodynamic polarization studies revealed that six leaves extract acts as mixed type inhibitor; they suppressed both cathodic and anodic process.Electrochemical impedance measurements showed the inhibitor adsorbs on the mild steel surface with increasing charge transfer resistance and decreasing of the double layer capacitance.The adsorption of the extract on the surface of mild steel follows Temkin adsorption isotherm.The adsorbed film over the mild steel surface has been confirmed by SEM analysis.

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