

Synergistic Effect of Zizyphus Spina-Christi and Chloride Ions on the Corrosion Inhibition of Mild Steel in Sulphuric Acid Solution.

Aisha M .Al-Turkustani^{*}, Sanaa T. Arab^{**} and Areej A. Al-Rehaili^{***}

^{*}King Abdulaziz University, Girls College of Education, Chemistry Department, Jeddah KSA
(A.M.Turkustani@hotmail.com),

^{**}King Abdulaziz University, Girls College of Education, Chemistry Department, Jeddah KSA (Prof.S.T.arab@hotmail.com).

^{***}Tabook University, Chemistry Department, Tabook KSA
(skooon_n-alil@hotmail.com)

ABSTRACT

The effect of chloride ions addition on the corrosion and corrosion inhibition of mild steel in 1.0 M H₂SO₄ containing 10% EtOH at 30°C have been investigated by using chemical and electrochemical measurements. The addition of chloride ions leads to stimulation of corrosion at $\leq (10^{-4} \text{ M})$ of chloride ions but, at concentrations $> (10^{-4} \text{ M})$ the increasing amount of chloride ions produces corrosion inhibition of mild steel. The calculated values of the synergistic parameter for the studied systems are near unity which indicates the occurrence of synergistic effect between the chloride ions and the studied extracts. The synergistic behavior of the chloride ions and extract components can be attributed to cooperative adsorption or competitive adsorption.

KEY WORDS: Synergistic, Corrosion inhibition, Zizyphus Spina-Chriti, Mild Steel, Sulphuric acid, Chloride ions.

1. INTRODUCTION

Corrosion is a fundamental process playing an important role in economics and safety, particularly for metals. The use of inhibitors is one of the most practical methods for protection against corrosion, especially in acidic media [1]. The use of corrosion inhibitors has considerably

increased in recent years due to increase awareness of corrosion worldwide. Inhibitors play an important role in controlling corrosion of metals [2].

Most corrosion inhibitors are known for their specificity of action. However, the combination of inhibitors is more likely to provide multiple effects required for effective corrosion inhibition. Interestingly, previous studies [3-13] have shown that addition of halide ions to acid/alkaline solution containing any organic compound has been found to result in synergistic effect, thereby inhibiting corrosion. The presence of halide ions in solution has been found to stabilize the adsorption of some organic cations leading to improved inhibition efficiency.

However, no reports exist, to the best of our knowledge, on the effect of chloride ion on the corrosion of mild steel in acidic environment using naturally occurring compounds such as *Zizyphus Spina-Christi* (ZSC). Therefore, in the present investigation, the synergistic effect of chloride ion on the performance of *Zizyphus Spina-Christi* (a natural occurring compounds), which is considered as non-toxic, inexpensive, environmentally friendly, and a renewable source of material as a corrosion inhibitor of mild steel in acidic (H_2SO_4) medium, is reported using chemical (weight loss and hydrogen evolution) and electrochemical (impedance and polarization) measurements at 30°C .

2. EXPERIMENTAL

2.1. Materials

The weight composition of the mild steel sample is the following: 0.275% Mn, 0.015% Ni, 0.004% Pb, 0.077% Al, and the remainder is iron (Fe). Before each experiment, the electrode was polished with different grade emery papers, degreased with acetone, rinsed by deionized water and finally dried with a stream of air.

2.2. Solutions

Stock solutions of the plant extracts (aqueous and/or alcoholic) were prepared by weight a certain amount (about 500 grams) of dried leaves of *Zizyphus Spina-Christi* plant and was crushed in an electric mixer, then add to it the appropriate solvent (deionized water and / or ethyl alcohol) and heat until boiling, the mixture is cooled for 24 hours and then be filtrated. Extraction is repeated several times from the same pool so extract collect and dried in the air to be concentrated and deposited to the least possible amount of solvent used. The extract was collect after concentrated it and placed in a standard flask 250 ml capacity and complete to the mark with the appropriate solvent and therefore is a solution under study.

The studied solutions were made of AR grade H_2SO_4 and KCl. One molar concentration of acid (1.0 M H_2SO_4) was prepared using deionized water. The concentrations of the inhibitor employed (extracts) were 0.2%v/v to 10%v/v. The concentrations of chloride ions are in the range ($1.0 \times 10^{-4} - 5.0 \times 10^{-2}$) M which used in the synergistic study.

2.3. Measurements

The chemical (hydrogen evolution (HE) and mass loss (ML)) measurements were carried

out as described elsewhere [14]. The electrochemical (electrochemical impedance spectroscopy (EIS) and potentiodynamic polarization (PDP)) measurements have been performed with an ACM Gill AC instruments relating to a computer. (Bridge DVD ASUS 8X max). For polarization and impedance studies, the steel specimen was with an exposed area of 0.949 cm² as a working electrode. A platinum foil was used as an auxiliary electrode. The reference electrode was a Ag / AgCl_(s) KCl_(sat.soln.) coupled to a Luggin capillary whose tip was located between the working electrode and the auxiliary electrode.

The inhibition efficiency from chemical methods (Inh.HE% and Inh.MS%) and electrochemical methods (Inh.EIS% and Inh.PDP%) were calculated from the following relations, respectively:

$$\text{Inh.HE\%} = (1 - R / R_o) \times 100 \quad (1)$$

$$\text{Inh.ML\%} = (1 - R' / R'_o) \times 100 \quad (2)$$

$$\text{Inh.R}_{ct} \% = (R_{cto}^{-1} - R_{ct}^{-1} / R_{cto}^{-1}) \times 100 \quad (3)$$

$$\text{Inh.p\%} = (1 - I_{corr} / I_{corr}^o) \times 100 \quad (4)$$

where, R , R_o , R' , R'_o , I_{corr} , I_{corr}^o , R_{ct} and R_{cto} are the corrosion rate, the corrosion current and the charge transfer resistance for steel in absence and presence of certain concentrations of **ZSC** extracts from HE, ML, EIS and PDP methods, respectively.

Studies have shown earlier that the chloride ion has the ability to adsorption on the surface of steel more than sulfate ion [15-17] and thus serve to attract the inhibitor by electrostatic forces, leading to increased layer protection cover the surface of the metal [18-20].

3.1. Effect of Chloride Ions on the Corrosion of Mild Steel at 30°C (Inhibition and Stimulating).

The corrosion for mild steel was studied in 1.0 M H₂SO₄ containing 10% EtOH in presence of different concentrations in the range (1.0×10^{-4} - 5.0×10^{-2}) M of potassium chloride (KCl) at 30°C using chemical and electrochemical methods.

3.1.1. Chemical Study:

Chemical measurements were conducted using hydrogen evolution (HE) and mass loss (ML) methods. Figure (1) shows the relation of hydrogen gas with time for mild steel corrosion in 1.0 M H₂SO₄ solution containing 10% EtOH in absence and presence of different concentrations of chloride ions at 30°C. The figure shows that, at the low concentration (1.0×10^{-4} M) of chloride ions acceleration of the corrosion occurs (increase in the slope of the curve), and the corrosion rate of the sample decrease by increasing the concentration of chloride ions i.e. increases the inhibition efficiency. Similar results were obtained from mass loss measurements. Table (1) illustrate the corrosion rates (R and R') and inhibition efficiency (Inh.%) obtained from chemical measurements (HE and ML), respectively.

3.1.2. Electrochemical Study:

Figures (2 and 3) and tables (2 and 3) give the electrochemical behavior (impedance and polarization) for mild steel corrosion in 1.0 M H₂SO₄ solution containing 10% EtOH in absence and presence of different concentrations of chloride ions at 30°C. Figure (2) illustrates Nyquist curves, which shows an increase in the charge transfer resistance (R_{ct}) (increase in diameter of semi-circles) with increasing Cl⁻ concentration, also the capacitive circuit are complete and closed. As appears from the figure a shift of the impedance at high frequency on the real axis of impedance and this may due to the electrolyte solution resistance R_s between working electrode and reference electrode [21].

$$R_s = 1 / K \pi r^2 \quad (5)$$

Where R_s solution resistance, r electrode diameter and K conductivity of electrolyte solution. At high frequencies, the real impedance are equivalent to only solution resistance, while at low frequencies, the real impedance are equivalent to solution

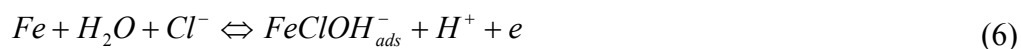
resistance and resistance of oxide layer of polarization ($R_s + R_p$) at which the value of the polarization resistance (R_p) depends on the electronic conductivity of the film layer and the rate of dissolution or the formation [22]. The electrical equivalent circuit of the pervious system give as follows [$R_s(C_{dl}R_{ct}(QR))$] at all concentrations, where C_{dl} electrical double layer capacity, R_{ct} charge transfer resistance and Q component of a phase constant.

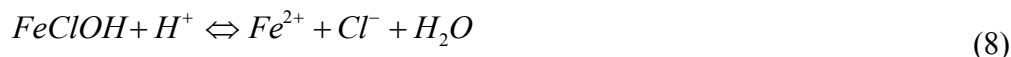
Table (2) shows that n values were close to unity at all concentrations which is evidence that the surface is smooth and homogenous. Also it is noted from the table, that the chloride ions gave an acceleration at low concentration (1.0×10^{-4}) M, but at high concentrations it gives an inhibition of corrosion which increase with increasing the concentration of chloride ions. The same behavior was obtained from polarization study, Figuer (3) and Table (3), where it is clear that chloride ions hinder both the cathodic and anodic process (metal dissolution and hydrogen evolution) as shown in the displacement of cathodic and anodic Tafel lines.

The results obtained from the chemical and electrochemical techniques can be explained as follows: chloride ions gave a dual action in the inhibition process is either accelerate or inhibit the process of corrosion based on its concentration in the solution this is consistent with the results reached in previous studies [23-26].

Increasing corrosion rate at low concentrations can be explained as that the chloride ions increase the metal dissolution by adsorption and it, concentrates in the anodic holes containing intermediate complex which working to increase the corrosion rate [27-29], it is found that chloride ions play a role in the degradation of the inert layer formed on the metal surface (interface between metal and solution) through the penetration of this layer and removing water molecules adsorb on it and then to accelerate the rate of corrosion [30]. At high concentrations the inhibitive action of chloride ions, a negative charge is attributed to their accumulation in the solution and adsorption easily on the metal surface positively charged [31,32] leading to the inhibition of metal corrosion in an acid environment [31], also the increase in chloride ion concentration lead to increase the ability of polarization, which stimulates the process of adsorption not only under the electrostatic influence but also through the formation of covalent bonds with the surface of mild steel which lead to more inhibition efficiency.

The iron dissolution process depends primarily on the intermediate complex and also depends on the type and nature of the ions in the corrosion environment, but in the case of the presence of chloride ions the dissolution mechanism as follows [33].





It is assumed that the dissolution of intermediate complex (equation (7)) depends on the concentration of chloride ions, it expected a decrease in solubility of the complex as increase in chloride ion concentration and this leads to more inhibition.

3.2. Corrosion Inhibition of Mild Steel in 1.0 M H₂SO₄ Containing 10% EtOH at Different Concentrations of Aqueous and Alcoholic Extracts of *zizyphus Spina-Christi* in Presence of 0.01 M Chloride Ions at 30 °C.

The corrosion of mild steel in 1.0 M H₂SO₄ containing 10% EtOH in the presence of different concentrations of aqueous extract (A) and alcoholic extract (B) of *Zizyphus Spina-Christi* plant and in presence of 0.01M of the chloride ions was studied using chemical and electrochemical methods to know the role played by chloride ion to promote or inhibit the corrosion of mild steel in acidic environment in the presence of extracts under study.

3.2.1. Chemical and Electrochemical Study:

Tables (4) gives the corrosion rates and the inhibition efficiencies calculated from the chemical methods for mild steel in presence of different concentrations of aqueous extract (A) and , alcoholic extract (B), respectively, and in presence of 0.01M chloride ions, where a decrease in the corrosion rate (increase in the inhibition efficiency) is found by increasing the concentration of inhibitor but in the case of alcoholic extract (B) the inhibition efficiency reached to 100% and 97.82% at a concentration 5% v/v, from HE and ML methods, respectively, then it decrease in a simple degree.

In the electrochemical study figure (4), Nyquist plots shows that in the case of aqueous extract the presence of capacitive circle with tail which increase in diameter accompanied by increasing the concentration of extract, especially at high concentrations of aqueous extract Fig. (4a), while in the case of alcoholic extract Fig. (4b) an increase in the capacity of the circle up to 7.0 % v/v after that the capacity increase by increasing the concentration of alcoholic extract. The electrochemical impedance parameters were recorded in Table (6) for both aqueous extract(A) and alcoholic extract (B), where it notes a decrease in the value of the electrical double layer capacity (C_{dl}) by increasing the concentration of the extract corresponding to an increase in the value of Charge Transfer resistance (R_{ct}), this leads to increase the inhibition. The simulation of electrochemical processes it is found that the equivalent circuit model proposed as follows:

- a-** at low concentrations (0.2 - 1.0)% v/v in the case of aqueous extract and at all concentrations of the alcoholic extract is: Rs[C_{dl}R_{ct}(QR)].
- b-** at the rest of high concentrations of aqueous extract and the concentration 0.5% v/v of the alcoholic extract the equivalent electrical circuit is: Rs[Q(R_{ct}(C_{dl}R))].)

In the potentiodynamic polarization study (PDP) (Figure 6) the effect of adding 0.01 M of chloride ions show that cathodic and anodic Tafel lines are displaced which indicates that no change on both cathodic and anodic processes. The electrochemical variables were recorded in Table (7) which indicates a decrease in the value of corrosion current (I_{corr}) when add both extracts, leading to increase the inhibition efficiency as the concentration of water and/or alcoholic extract is increased in the presence of 0.01M chloride ions. This result agreed with the conclusions obtained from chemical (HEM and MLM) and impedance (EIS) studies, so the added 0.01 M of chloride ions enhances the inhibition effectiveness of the studied extracts.

3.3. Study of Synergistic and Antagonism Effect of Chloride Ions in Acidic Solutions:

The synergistic effect between chloride ions and cations of the extract can be attributed to a cooperative adsorption, which called co-adsorption, also competitive adsorptions due to competition between different ions (anions and cations) in solution, if the measured value Θ'_{A+C} greater than the calculated value Θ_{A+C} , then the synergistic effect is due to the cooperative adsorption on the metal surface, which occurs as a result of electrostatic attraction between each of the negative chloride ion and positive extracted cation, whereas if the measured value is less than the calculated value the synergistic effect is due to the competitive adsorption between different ions existing in the solution [34].

Where Θ_A represents the anion surface coverage, Θ_C the cation surface coverage, Θ'_{A+B} the measured surface coverage for the anion and cation together and Θ_{A+C} the calculated surface coverage for the anion and cation together which it calculate from the following relationship:

$$\Theta_{A+C} = (\Theta_A + \Theta_C) - (\Theta_A \Theta_C) \quad (9)$$

The values of synergistic coefficient S_Θ were calculated using the relationship [35]:

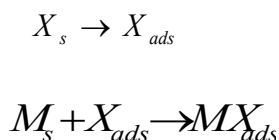
$$S_\Theta = \frac{1 - \Theta_{(A+C)}}{1 - \Theta'_{(A+C)}} \quad (10)$$

If S_Θ values greater than unity this indicates the existence of synergistic effect between the molecules of inhibitor (ZSC) and halide ion (chloride ion). If S_Θ values were less than unity this indicate on the antagonism effect [38]. Tables (8 and 9) represent the values of synergistic coefficient S_Θ and values of the surface coverage Θ derived from electrochemical (impedance and polarization) measurements for the corrosion of mild steel in 1.0 M H_2SO_4 containing 10% EtOH at different concentrations of aqueous extract

and alcoholic extract of ZSC plant in the presence of 0.01M chloride ions from where they can discuss the results as follows:

Note that the values of S_θ from electrochemical methods for both extracts (aqueous extract and alcoholic extract) greater than unity and at some concentrations are less than unity this indicating to increase the inhibition of mild steel corrosion by adding chloride ions to the extracts studied, this is due to an synergistic effect between the molecules of the extracts and chloride ions [36,37]. Comparing the values in Tables (8 and 9) illustrate the existence of both types of adsorption synergistic and competitive.

Synergistic effect can be explained by that the chloride ions are first chemically adsorb on the mild steel surface of which facilitate the adsorption process of the molecules of water extract and / or alcoholic extract by attraction by Coulomb forces (different charges) which contain complex compound working to increase the coverage of the surface of mild steel under study and thus increase the inhibition efficiency, the mechanism of interaction has been proposed as follow [38]:



Where X_s and M_s refer to chloride ions and extracted molecules, respectively, X_{ads} and MX_{ads} refer to chloride ions and ionic compound, respectively, in the case of adsorption.

3.4. Corrosion Inhibition of Mild Steel in 1.0 M H₂SO₄ Containing 10% EtOH at 0.5% v/v of the Two Studied Extracts at different Concentrations of Chloride Ions.

The effect of adding different concentrations of potassium chloride in the range (1.0×10^{-4} - 5.0×10^{-2}) M on the inhibition of mild steel corrosion in 1.0M H₂SO₄ solution containing 10% EtOH in the presence of constant concentration (0.5% v/v) of aqueous extract and/or alcoholic extract of ZSC plant using chemical and electrochemical methods has been studied.

Table (10), shows a low corrosion rate than in the case of acid free and the inhibition efficiency increases with increasing chloride ions concentration, this behavior is clear in the case of alcoholic extract, which give inhibition 100% and 96.03% from chemical study and 81.80% and 83.54% in the case of aqueous extract, respectively, this means that the alcoholic extract of ZSC plant is more effective than the aqueous extract in the presence of different concentrations of chloride ions. It also shown from the two tables

that at low concentrations of chloride ions (1.0×10^{-4} - 5.0×10^{-3}) M the inhibition efficiency were lower than those in the case the extracts only, this possibly due to the interaction between chloride ions and mild steel surface which result an increase the corrosion rate and reduce the inhibition efficiency, this confirm the dual behavior of the chloride ion in acidic solution, as mentioned earlier [23-26].

In the electrochemical study, Figure (6) illustrates Nyquist plots that the impedance curves often give half circle indicates that the process of corrosion are mainly controlled by the charge transfer process and the apparent difference is due to the current dispersion. The variables of impedance are recorded in Table (11) where it was noted that : in general, an increase in the value of the charge transfer resistance this means increase the inhibition efficiency with increasing the concentration of chloride ions (the alcoholic extract give higher efficiency than the aqueous extract) as can be seen from the table and also noted that low concentrations of chloride ions gives the inhibition efficiency less than those in the case of the use of aqueous and alcoholic extracts only, and when simulate the electrochemical processes it is found that the type of electrical circuit in the case of aqueous extract at all concentrations is: $R[CR(QR)]$, and in the case of alcoholic extract the electrical circuit at all concentrations of the type: $R[C(QR)]$, with the exception of concentration 5.0×10^{-2} M the type of the circuit is: $R[Q(CR)]$, it also notes that the values of n observed in the Table were close to unit, which indicates that the capacity loops are closed and the surface is smooth and in the case of alcoholic extract at the highest concentration of chloride ions the value of n is equal to 0.296, this indicates that the capacity loop is open and it accompanied with spread tail as shown in Figure (6).

Figure (7) shows polarization curves, it is clear from the figure that the presence of both extracts with different concentrations of chloride ions hinder both anodic and cathodic processes which appear from the shift in Tafel lines to anodic and cathodic directions which means that both two extracts act as mixed inhibitors in the presence of chloride ions. The electrochemical parameters are recorded in Table (12), which shows that the results is supported that results obtained from the impedance measurements, which indicates a decrease in corrosion current with increasing the chloride ions concentration and thus increase the inhibition efficiency, especially in the case of alcoholic extract which gives inhibition efficiency higher than that for aqueous extract, noting that the decrease in inhibition efficiency than those in the case of extracts alone in the presence of low concentrations of chloride ions and it was noted that this behavior is similar to the behavior from chemical measurements and the results are interpreted as follows:

The increase in the inhibition efficiencies for the aqueous extract and alcoholic extract with increasing the concentration of chloride ions is attributed to an electrostatic

attraction between chloride ions and the adsorbed components of the extract which lead to the formation of ionic complex MX_{ads} between the components extracted and chloride ion adsorb on mild steel surface, leading to increased the surface coverage and enhance the inhibition process [39].

The mechanism of inhibitor adsorption in the presence of halide ions has been interpreted by Mc Cafferty [40] as follows:

- The competitive with halide ions on the position of water molecules covering the metal surface, where the inhibitor molecules loss the positive proton through electrical double layer and adsorbed it on the metal surface through the free pair of electrons.
- Adsorption of the positive inhibitor molecules on halide covering the surface through the nitrogen or oxygen atom. It was noted that both the previous mechanism can increase the inhibition of corrosion.

The decrease in inhibition efficiency at low concentrations of chloride ions due to the removal of particles from the extract which adsorb on the metal surface by chloride ions [41] where it react with the metal surface, which results an increase in the corrosion rate of mild steel and therefore reducing the inhibition efficiency.

Tables (13 and 14) illustrates the values of synergistic coefficients and surface coverage of mild steel corrosion in the presence of different concentrations of chloride ions and constant concentration of the aqueous extract and/or alcoholic extract, respectively from electrochemical measurements. It is clear from both Tables that the values of synergistic coefficient at low concentrations of chloride ions is less than unity, and at high concentrations of chloride ions it gives values close to unity due to the competitive adsorption between the chloride ions and molecules of aqueous and alcoholic extracts.

CONCLUSIONS

From the chemical and electrochemical studies, it can be easily concluded that as follows:

The addition of chloride ions leads to stimulation of corrosion at $\leq (10^{-4} \text{ M})$ of chloride ions out at concentration $> (10^{-4} \text{ M})$ of chloride ions increasing amount of chloride ions produces corrosion inhibition of mild steel, will occurred :

- * The addition of 0.01 M chloride ions enhances the inhibition efficiency of *Zizyphus Spina-Christi* extracts.
- * The addition of increasing amount of chloride ions in the presence of certain concentration of *zizyphus Spina-Christi* extracts leads to increase the inhibitor efficiency and the calculated values of the synergistic parameter for the studied

systems are near unity which indicates the occurrence of synergistic effect between the chloride ions and the studied extract.

- * The synergistic behavior the chloride ions and extract components be attributed to cooperative adsorption or competitive adsorption.

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Table (1): Corrosion rate and Inh.% for mild steel corrosion in 1.0 M H₂SO₄ + 10% EtOH in presence of different concentrations of Cl⁻ ions at 30°C.

C_{Cl^-} (M)	Corrosion Rate		Inhibition Efficiency	
	$R_{HEM} \times 10^2$ (ml. cm. ⁻² min. ⁻¹)		$R'_{MLM} \times 10^3$ (g. cm. ⁻² min. ⁻¹)	
0.0	3.704	7.256	-	-
1.0×10^{-4}	3.870	7.477	-4.50	-2.76
2.5×10^{-3}	3.106	6.120	16.12	15.66
5.0×10^{-3}	2.498	5.513	32.56	28.98
1.0×10^{-2}	2.179	4.475	41.17	38.32
2.5×10^{-2}	1.999	4.097	46.02	43.54
5.0×10^{-2}	1.412	3.569	61.88	50.82

Table (2): Electrochemical impedance variables for mild steel corrosion in 1.0 M H₂SO₄ + 10% EtOH in absence and presence of different concentrations of chloride ions at 30°C.

C_{Cl^-} (M)	$R_{sol.}$ (Ω .cm. ⁻²)	C_{dl} (μ F.cm. ⁻²)	R_{ct} (Ω .cm. ⁻²)	Q (Ω^{-1} .cm. ⁻² s ⁿ)	n	Inh.% (R_{cl}^{-1})
0.0	1.007	163.6	4.521	2.358×10^{-3}	0.948	-
1.0×10^{-4}	1.460	175.7	3.611	4.809×10^{-4}	0.909	-25.20
2.5×10^{-3}	1.551	177.4	5.144	2.584×10^{-4}	0.875	12.11
5.0×10^{-3}	1.490	119.0	5.348	1.210×10^{-2}	0.993	15.46
1.0×10^{-2}	1.338	137.9	5.619	3.155×10^{-2}	0.930	19.54
2.5×10^{-2}	1.249	108.9	7.550	1.174×10^{-3}	0.894	40.11
5.0×10^{-2}	1.826	118.6	7.935	1.280×10^{-4}	0.988	43.02

Table (3): Polarization parameters for mild steel corrosion in 1.0 M H₂SO₄ + 10% EtOH in absence and presence of different concentrations of chloride ions at 30°C.

C_{Cl^-} (M)	$-E_{corr}$ (v)	$I_{corr.}$ (mA.cm. ⁻²)	β_a (mV.dec. ⁻¹)	$-\beta_c$ (mV.dec. ⁻¹)	Inh.%
0.0	0.440	15.17	40.10	86.80	-
1.0×10^{-4}	0.440	16.18	71.42	171.4	-6.65
2.5×10^{-3}	0.451	13.10	50.94	116.4	13.64
5.0×10^{-3}	0.444	12.15	63.50	136.2	19.90
1.0×10^{-2}	0.440	12.00	58.33	107.1	20.89
2.5×10^{-2}	0.447	8.880	50.69	82.99	41.47
5.0×10^{-2}	0.445	6.755	54.32	83.15	55.48

Table (4): Corrosion rates and inhibition efficiencies for mild steel in 1.0 M H₂SO₄ +10% EtOH in presence of different concentrations of aqueous extract of Zizyphus Spina Christi plant and in presence 0.01 M of chloride ions 30°C.

C_{inh} (V/V%)	Corrosion Rate		Inhibition Efficiency	
	$R_{HEM} \times 10^2$ (ml. cm. ⁻² min. ⁻¹)	$R'_{MLM} \times 10^5$ (g. cm. ⁻² min. ⁻¹)	$Inh_{HEM}\%$	$Inh_{MLM}\%$
0.0	3.704	7.256	-	-
0.2	2.507	4.891	32.32	32.59
0.5	1.991	4.900	46.24	42.25
1.0	1.003	2.938	72.92	59.50
2.0	0.840	2.330	77.31	67.88
10.0	0.431	1.093	88.35	84.91
16.0	0.295	0.621	92.03	91.42
20.0	0.097	0.016	97.35	91.70

Table (5): Corrosion rates and inhibition efficiencies for mild steel in 1.0 M H₂SO₄ +10% EtOH in presence of different concentrations of alcoholic extract of Zizyphus Spina Christi plant and in presence 0.01 M of chloride ions 30°C.

C_{inh} (V/V%)	Corrosion Rate		Inhibition Efficiency	
	$R_{HEM} \times 10^2$ (ml. cm. ⁻² min. ⁻¹)	$R'_{MLM} \times 10^5$ (g. cm. ⁻² min. ⁻¹)	$Inh_{HEM}\%$	$Inh_{MLM}\%$
0.0	3.704	7.256	-	-
0.2	0.736	1.007	80.11	86.11
0.5	0.150	0.704	95.92	90.29
2.0	0.00	0.223	100	96.92
5.0	0.00	0.158	100	97.82
7.0	0.00	0.195	100	97.30
10.0	0.00	0.197	100	97.28

Table (6): Electrochemical impedance variables for mild steel corrosion in 1.0 M H₂SO₄ + 10% EtOH in the presence of different concentrations of aqueous extract (A) and alcoholic extract (B) of the plant and 0.01M of the chloride ions at 30°C.

	C_{Ex} (V/V%)	$R_{sol.}$ ($\Omega.cm.^{-2}$)	C_{dl} ($\mu F.cm.^{-2}$)	R_{ct} ($\Omega.cm.^2$)	Q ($\Omega.^{-1}cm.^{-2}s^n$)	n	$Inh.\%$ (R_{ct}^{-1})
Extract (A)	0.0	1.007	163.6	4.521	2.358×10^{-3}	0.948	-
	0.2	1.181	102.6	6.720	1.422×10^{-4}	0.924	32.72
	0.5	1.664	105.7	8.385	3.282×10^{-3}	0.839	46.08
	1.0	2.090	75.86	9.681	3.020×10^{-3}	0.757	53.30
	2.0	1.607	66.75	12.87	0.485×10^{-3}	0.639	64.87
	10.0	0.702	46.40	20.26	9.547×10^{-3}	0.800	77.68
	16.0	1.108	43.96	28.84	6.743×10^{-3}	0.800	84.32
	20.0	0.806	41.19	28.89	2.386×10^{-3}	0.265	84.35
Extract (B)	0.2	2.571	60.50	13.76	2.609×10^{-5}	0.770	67.14
	0.5	0.781	45.87	29.10	4.539×10^{-3}	0.800	81.24
	2.0	2.893	28.76	57.37	6.276×10^{-5}	0.627	92.11
	5.0	3.184	24.42	86.27	5.073×10^{-5}	0.874	94.75
	7.0	2.703	22.26	91.41	4.207×10^{-5}	0.420	95.05
	10.0	2.770	22.83	86.52	5.416×10^{-6}	0.860	94.77

Table (7) : Variables obtained from polarization for mild steel corrosion in 1.0 M H₂SO₄ + 10% EtOH in the presence of different concentrations of aqueous extract (A) and alcoholic extract (B) of the ZSC plant and 0.01 M of chloride ions at 30 ° C.

	C_{Ex} (V/V%)	$-E_{corr.}$ (v)	$I_{corr.}$ (mA.cm. ⁻²)	β_a (mV.dec. ⁻¹)	$-\beta_c$ (mV.dec. ⁻¹)	$Inh.\%$
Extract (A)	0.0	0.450	15.17	40.10	86.80	-
	0.2	0.459	10.45	48.15	88.20	30.52
	0.5	0.431	6.390	38.93	75.23	57.85
	1.0	0.447	6.205	43.45	81.08	59.09
	2.0	0.450	5.912	69.98	140.9	61.02
	10.0	0.439	5.03	34.70	75.21	66.84
	16.0	0.451	3.814	52.05	92.21	74.88
	20.0	0.451	3.757	41.58	70.36	75.23
Extract (B)	0.2	0.461	4.85	55.20	98.65	68.02
	0.5	0.452	4.61	34.29	50.58	69.55
	2.0	0.454	3.59	30.59	49.55	76.55
	5.0	0.451	1.89	27.40	36.69	87.49
	7.0	0.460	2.84	33.18	42.36	81.27
	10.0	0.445	3.02	35.53	68.68	80.09

Table (8): Synergistic coefficient and surface coverage values for mild steel corrosion in 1.0 M H₂SO₄ + 10% EtOH in presence of different concentrations of aqueous extract and 0.01 M chloride ions from electrochemical measurements at 30 °C.

Method	$C_{Ex.}$ (V/V)%	C_{Cl^-} (M)	θ_A	θ_C	$\theta_{(A+C)}$ (calculate d)	$\theta'_{(A+C)}$ (measure d)	S_θ
Impedance	0.01	0.1954
	0.2	0.3102
	0.2	0.01	0.4449	0.3272	0.82
	0.5	0.4722
	0.5	0.01	0.5753	0.4608	0.80
	1.0	0.5359
	1.0	0.01	0.6265	0.5330	0.80
	2.0	0.5640
	2.0	0.01	0.6326	0.6487	1.04
	10.0	0.6926
	10.0	0.01	0.7526	0.7768	1.10
	16.0	0.7651
	16.0	0.01	0.8109	0.8432	1.00
	20.0	0.8051
	20.0	0.01	0.8431	0.8435	1.20
Polarization	0.01	0.2089
	0.2	0.2817
	0.2	0.01	0.4317	0.3052	0.81
	0.5	0.3805
	0.5	0.01	0.5099	0.5785	1.16
	1.0	0.4471
	1.0	0.01	0.5626	0.5909	1.06
	2.0	0.4813
	2.0	0.01	0.5896	0.6102	1.05
	10.0	0.5453
	10.0	0.01	0.6402	0.6684	1.08
	16.0	0.6357
	16.0	0.01	0.7118	0.7488	1.14
	20.0	0.6406
	20.0	0.01	0.7158	0.7523	1.14

Table (9): Synergistic coefficient and surface coverage values for mild steel corrosion in 1.0 M H₂SO₄ + 10% EtOH in presence of different concentrations of alcoholic extract and 0.01 M chloride ions from electrochemical measurements at 30 °C.

Method	$C_{Ex.}$ (V/V)%	C_{Cl^-} (M)	θ_A	θ_C	$\theta_{(A+C)}(calculated)$	$\theta'_{(A+C)}(measured)$	S_0
Impedance	0.01	0.1954
	0.2	0.5915
	0.2	0.01	0.6557	0.6714	1.04
	0.5	0.7599
	0.5	0.01	0.8068	0.8124	1.02
	2.0	0.8604
	2.0	0.01	0.8876	0.9211	1.42
	5.0	0.8719
	5.0	0.01	0.8969	0.9475	1.96
	7.0	0.9243
	7.0	0.01	0.9390	0.9505	1.23
Polarization	10.0	0.9070
	10.0	0.01	0.9172	0.9477	1.58
	0.01	0.2089
	0.2	0.5324
	0.2	0.01	0.6300	0.6802	1.15
	0.5	0.5400
	0.5	0.01	0.6360	0.6955	1.19
	2.0	0.7614
	2.0	0.01	0.8112	0.7686	0.81
	5.0	0.8711
	5.0	0.01	0.8980	0.8749	0.81
	7.0	0.7996
	7.0	0.01	0.8414	0.8127	0.84
	10.0	0.7867
	10.0	0.01	0.8312	0.8009	0.84

Table (10): The corrosion rates and inhibition efficiencies of mild steel corrosion in 1.0 M H₂SO₄ + 10% EtOH in the presence of different concentrations of chloride ions containing 0.5% v / v of aqueous extract(A) and alcoholic extract (B) of the RC plant at 30 °C.

	$C_{Cl^-}(M)$	Corrosion Rate		Inhibition Efficiency	
		$R_{HEM} \times 10^2$ (ml. cm. ⁻² min. ⁻¹)	$R'_{MLM} \times 10^5$ (g. cm. ⁻² min. ⁻¹)	$Inh_{HEM}\%$	$Inh_{MLM}\%$
Extract(A)	0.0	3.704	7.256	-	-
	1.0×10^{-4}	3.018	5.995	18.50	14.19
	2.5×10^{-3}	2.916	5.838	21.25	19.53
	5.0×10^{-3}	2.743	5.296	25.92	27.01
	1.0×10^{-2}	1.991	4.900	46.24	42.25
	2.5×10^{-2}	1.308	2.686	64.66	62.98
	5.0×10^{-2}	0.673	1.194	81.80	83.54
Extract(B)	0.0	3.704	7.256	-	-
	1.0×10^{-4}	0.889	1.933	76.51	73.35
	2.5×10^{-3}	1.058	1.992	71.44	72.54
	5.0×10^{-3}	0.491	1.176	86.73	83.78
	1.0×10^{-2}	0.150	0.704	95.92	90.29
	2.5×10^{-2}	0.00	0.405	100.0	94.40
	5.0×10^{-2}	0.00	0.288	100.0	96.03

Table(11): Electrochemical impedance variables for mild steel corrosion in 1.0 M H₂SO₄ + 10% EtOH in the presence of 0.5% v / v of aqueous extract and/ or alcoholic extract of RC plant containing different concentrations of chloride ions at 30°C.

	C_{Ex} (V/V%)	R_{sol} ($\Omega.cm.^{-2}$)	C_{dl} ($\mu F.cm.^{-2}$)	R_{ct} ($\Omega.cm.^{-2}$)	Q ($\Omega^{-1}cm^{-2}s^n$)	n	$Inh.\%$ (R_{ct}^{-1})
Extract (A)	0.0	1.007	163.6	4.521	2.358×10^{-3}	0.948	-
	1.0×10^{-4}	1.453	120.9	4.606	1.214×10^{-4}	1.000	1.84
	2.5×10^{-3}	1.167	97.28	6.594	1.648×10^{-4}	0.859	31.43
	5.0×10^{-3}	2.228	89.04	7.350	4.076×10^{-4}	0.728	38.48
	1.0×10^{-2}	1.644	105.7	8.385	3.282×10^{-3}	0.839	46.08
	2.5×10^{-2}	1.272	94.45	8.509	1.815×10^{-4}	0.885	46.86
	5.0×10^{-2}	1.428	90.46	10.48	2.639×10^{-3}	0.831	56.86
Extract (B)	1.0×10^{-4}	1.762	73.25	10.54	1.929×10^{-3}	0.905	57.10
	2.5×10^{-3}	1.635	84.89	9.650	1.133×10^{-4}	0.950	53.15
	5.0×10^{-3}	2.314	56.13	17.09	5.506×10^{-5}	0.995	73.54
	1.0×10^{-2}	0.781	45.87	29.10	4.539×10^{-3}	0.800	81.24
	2.5×10^{-2}	2.243	36.69	33.23	7.575×10^{-5}	0.856	86.39
	5.0×10^{-2}	1.462	37.53	39.46	1.694×10^{-4}	0.296	88.54

Table(12): Polarization parameters for mild steel corrosion in 1.0 M H₂SO₄ + 10% EtOH in the presence of 0.5% v / v of aqueous extract (A) and alcoholic extract (B) of RC plant containing different concentrations of chloride ions at 30°C.

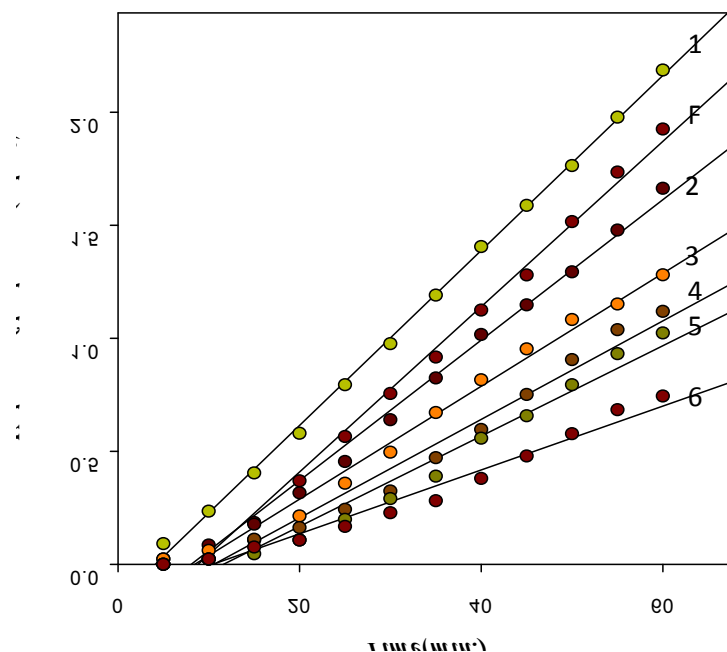
	C_{Ex} (V/V%)	$-E_{corr.}$ (v)	$I_{corr.}$ (mA.cm. ⁻²)	β_a (mV.dec. ⁻¹)	$-\beta_c$ (mV.dec. ⁻¹)	$Inh.\%$
Extract (A)	0.0	0.450	15.17	40.10	86.80	-
	1.0×10^{-4}	0.445	11.36	77.12	178.0	27.87
	2.5×10^{-3}	0.439	10.19	56.17	141.3	32.82
	5.0×10^{-3}	0.449	7.535	63.40	119.6	50.32
	1.0×10^{-2}	0.431	6.390	38.93	75.23	57.85
	2.5×10^{-2}	0.441	6.073	45.95	75.34	60.00
	5.0×10^{-2}	0.456	5.213	51.80	74.33	65.63
Extract (B)	1.0×10^{-4}	0.452	7.31	46.37	85.98	51.78
	2.5×10^{-3}	0.446	7.50	47.31	88.95	50.56
	5.0×10^{-3}	0.455	6.96	47.75	96.91	54.11
	1.0×10^{-2}	0.452	4.61	34.29	50.58	69.55
	2.5×10^{-2}	0.453	4.41	36.86	52.27	70.89
	5.0×10^{-2}	0.448	4.32	38.82	55.92	71.51

Table (13): Values of synergistic coefficient and surface coverage for mild steel corrosion in 1.0 M H₂SO₄ +10% EtOH and in presence 0.5%v/v of aqueous extract containing different concentrations of chloride ions at 30 ° C from electrochemical measurements.

<i>Method</i>	<i>C_{Cl⁻}</i> <i>(M)</i>	<i>C_{Ex.}</i> <i>(V/V)%</i>	<i>Θ_A</i>	<i>Θ_C</i>	<i>Θ_(A+C)(calculated)</i>	<i>Θ' _(A+C)(measured)</i>	<i>S_θ</i>
<i>Impedance</i>	0.5	0.4722
	1.0×10 ⁻⁴	-0.2520
	1.0×10 ⁻⁴	0.5	0.3391	0.0184	0.67
	2.5×10 ⁻³	0.1211
	2.5×10 ⁻³	0.5	0.5359	0.3143	0.67
	5.0×10 ⁻³	0.1546
	5.0×10 ⁻³	0.5	0.5537	0.3848	0.72
	1.0×10 ⁻²	0.1954
	1.0×10 ⁻²	0.5	0.5753	0.4608	0.78
	2.5×10 ⁻²	0.4011
<i>Polarization</i>	2.5×10 ⁻²	0.5	0.6839	0.4686	0.59
	5.0×10 ⁻²	0.4302
	5.0×10 ⁻²	0.5	0.6992	0.5686	0.69
	0.5	0.3805
	1.0×10 ⁻⁴	-0.0665
	1.0×10 ⁻⁴	0.5	0.3393	0.2787	0.91
	2.5×10 ⁻³	0.1364
	2.5×10 ⁻³	0.5	0.4649	0.3282	0.79
	5.0×10 ⁻³	0.1990
	5.0×10 ⁻³	0.5	0.5037	0.5032	0.99
	1.0×10 ⁻²	0.2089
	1.0×10 ⁻²	0.5	0.5099	0.5785	1.16
	2.5×10 ⁻²	0.4147
	2.5×10 ⁻²	0.5	0.6374	0.6000	0.90
	5.0×10 ⁻²	0.5548
	5.0×10 ⁻²	0.5	0.7241	0.6563	0.80

Table (14): Values of synergistic coefficient and surface coverage for mild steel corrosion in 1.0 M H₂SO₄ +10% EtOH and in presence 0.5% v/v of alcoholic extract containing different concentrations of chloride ions at 30°C from electrochemical measurements.

Method	C_{Cl^-} (M)	$C_{Ex.}$ (V/V)%	θ_A	θ_C	$\theta_{(A+C)}$ (calculated)	$\theta'_{(A+C)}$ (measured)	S_θ
Impedance	0.5	0.7599
	1.0×10^{-4}	-0.2520
	1.0×10^{-4}	0.5	0.6993	0.5710	0.70
	2.5×10^{-3}	0.1211
	2.5×10^{-3}	0.5	0.7889	0.5315	0.45
	5.0×10^{-3}	0.1546
	5.0×10^{-3}	0.5	0.7970	0.7354	0.76
	1.0×10^{-2}	0.1954
	1.0×10^{-2}	0.5	0.8068	0.8124	1.02
	2.5×10^{-2}	0.4011
	2.5×10^{-2}	0.5	0.8562	0.8639	1.05
	5.0×10^{-2}	0.4302
	5.0×10^{-2}	0.5	0.8631	0.8854	1.19
Polarization	0.5	0.5400
	1.0×10^{-4}	-0.0665
	1.0×10^{-4}	0.5	0.5094	0.5178	1.01
	2.5×10^{-3}	0.1364
	2.5×10^{-3}	0.5	0.6027	0.5056	0.80
	5.0×10^{-3}	0.1990
	5.0×10^{-3}	0.5	0.6315	0.5411	0.80
	1.0×10^{-2}	0.2089
	1.0×10^{-2}	0.5	0.6360	0.6955	1.19
	2.5×10^{-2}	0.4147
	2.5×10^{-2}	0.5	0.7307	0.7089	0.90
	5.0×10^{-2}	0.5548
	5.0×10^{-2}	0.5	0.7952	0.7151	0.71



[(F)Free, (1) 1×10^{-4} , (2) 2.5×10^{-3} , (3) 5×10^{-3} , (4) 1×10^{-2} , (5) 2.5×10^{-2} and (6) 5×10^{-2}] M

Fig.(1): The relation between hydrogen gas with time for mild steel corrosion in 1.0 M H_2SO_4 +10% EtOH in absence and presence of different concentrations of chloride ions at 30°C.

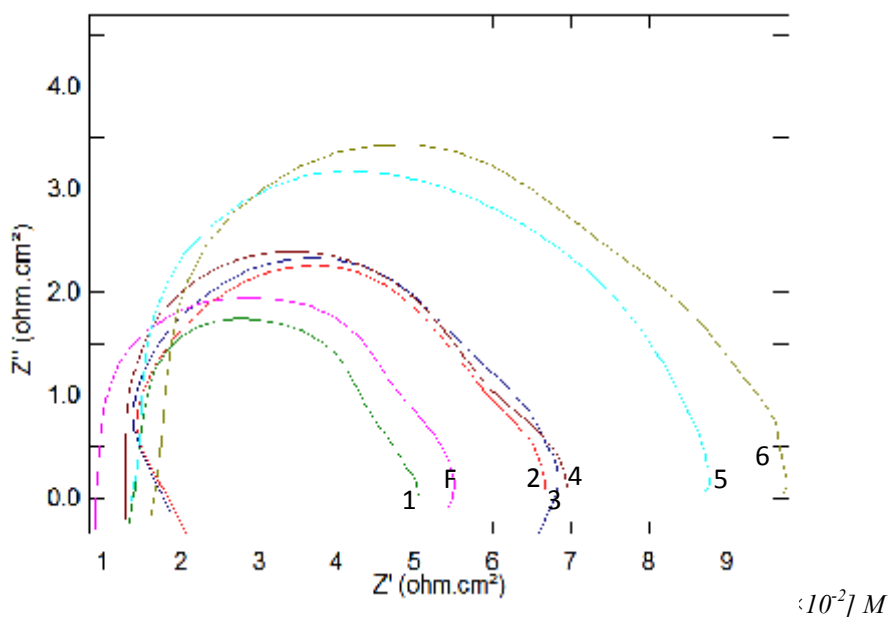
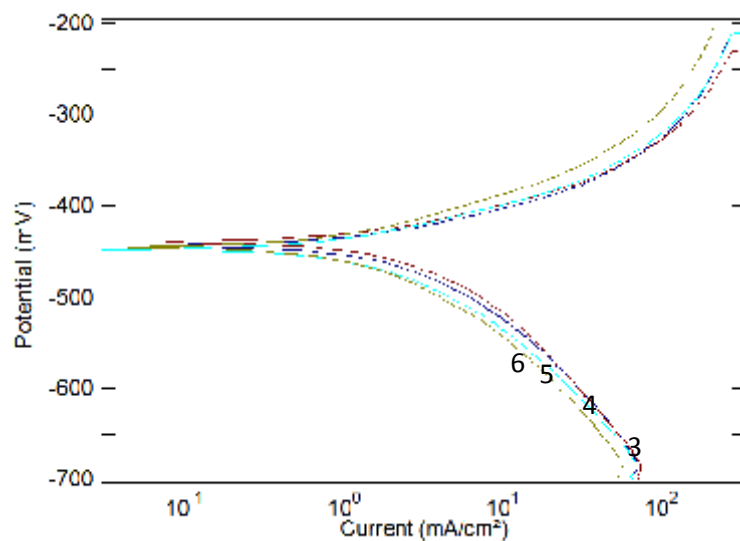
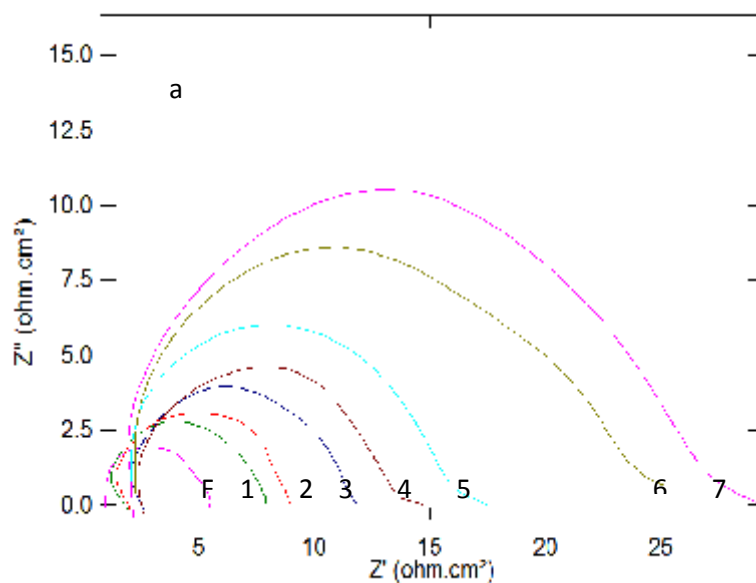


Fig. (2): Nyquist plots for mild steel corrosion in 1.0 M H_2SO_4 + 10% EtOH
in different concentrations of chloride ions at 30°C.

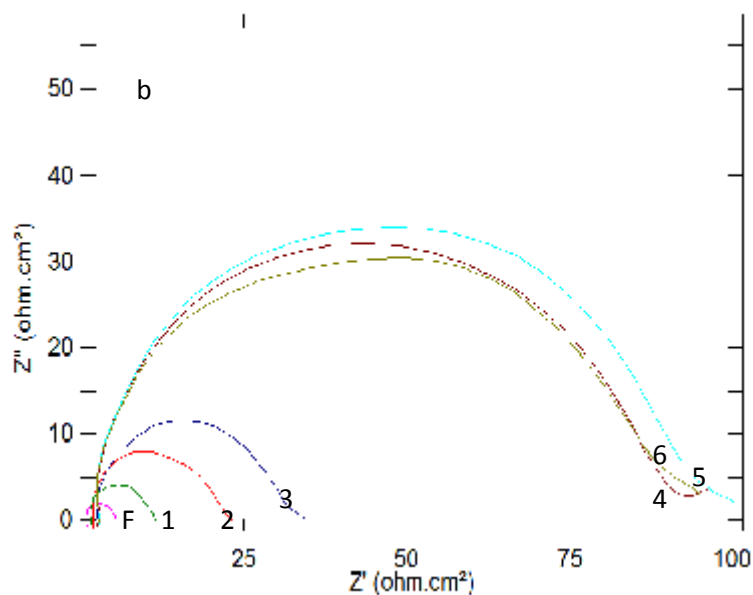


[(F)Free, (1) 1×10^{-4} , (2) 2.5×10^{-3} , (3) 5×10^{-3} , (4) 1×10^{-2} , (5) 2.5×10^{-2} and (6) 5×10^{-2}] M

Fig. (3): Polarization curves for mild steel corrosion in 1.0 M H_2SO_4 +
10% EtOH in different concentrations of chloride ions at 30°C.

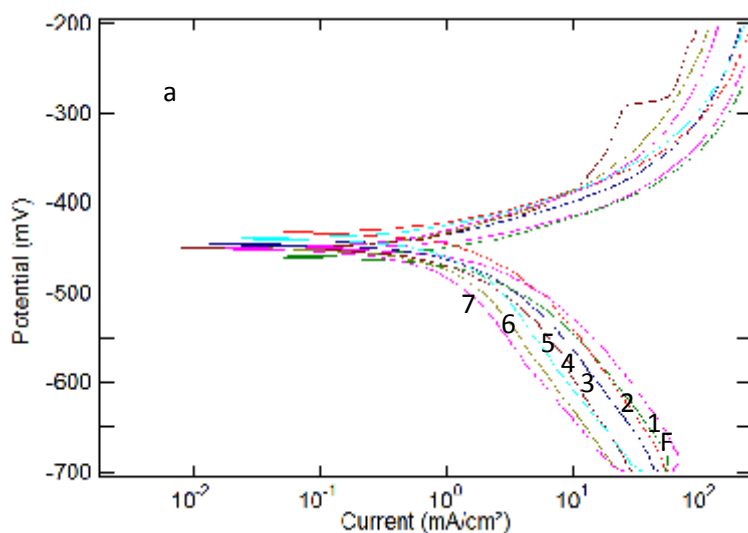


[(F)Free, (1)0.2, (2)0.5, (3)1.0, (4)2.0, (5)10.0, (6)16.0 and (7)20.0.] %v/v

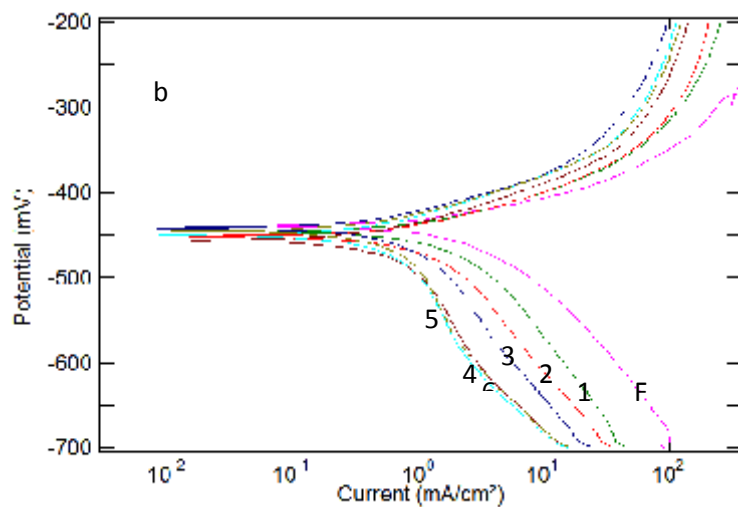


[(F) Free, (1)0.2, (2)0.5, (3) 2.0, (4)5.0, (5) 7.0 and (6)10.0]% v/v

Fig.(4): Nyquist plots for mild steel corrosion in 1.0 M H₂SO₄ +10% EtOH in presence of different concentrations of (a) aqueous extract (b) alcoholic extract and 0.01 M chloride ions at 30°C.

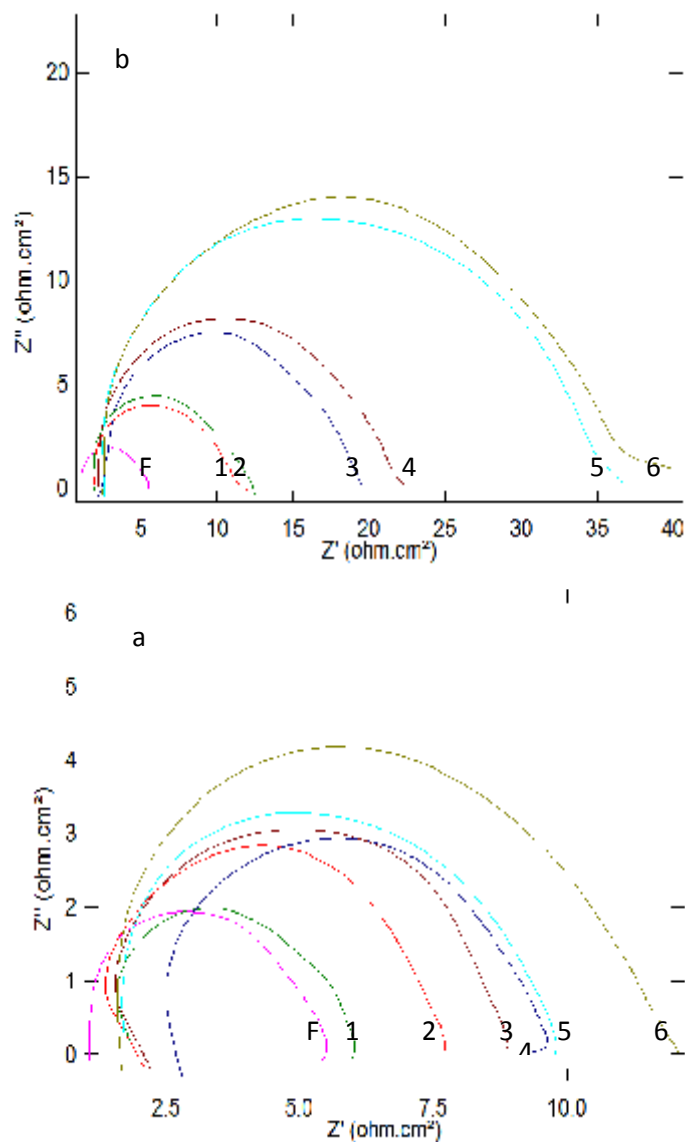


[(F)Free, (1)0.2, (2)0.5, (3)1.0, (4)2.0, (5)10.0(6)16.0 and (7)20.0.]%v/v



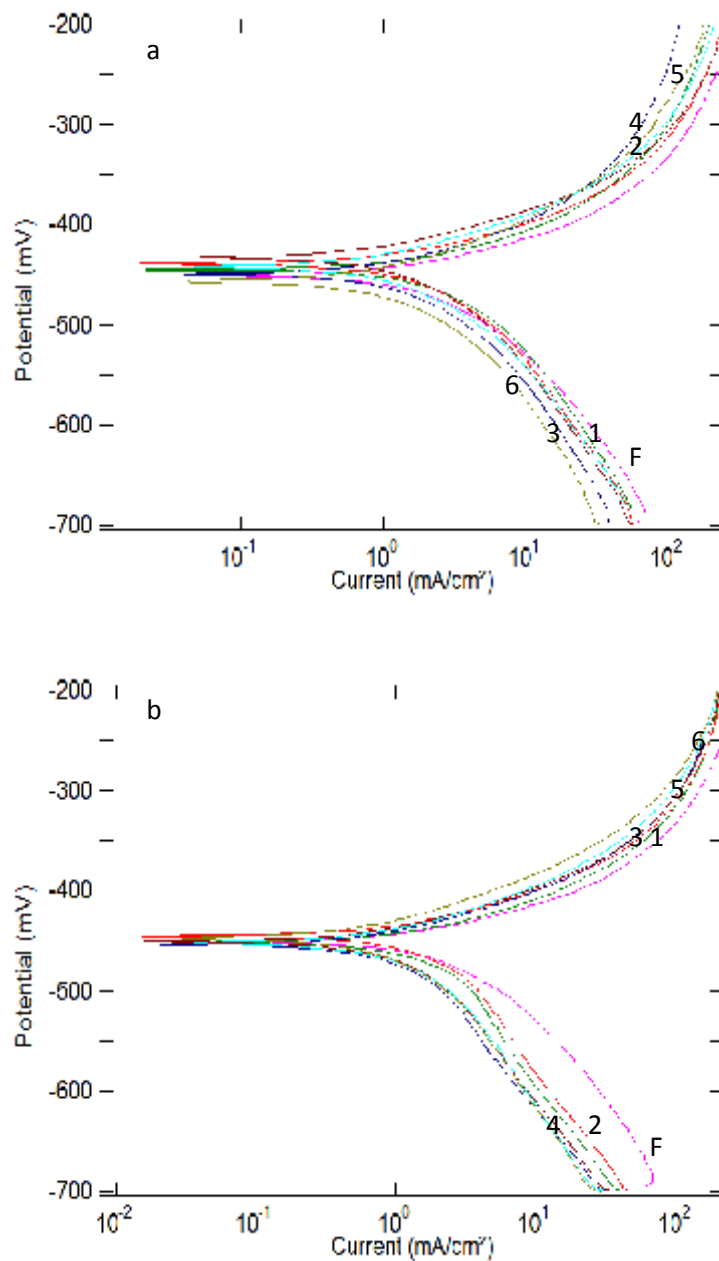
[(F) Free, (1)0.2, (2)0.5, (3) 2.0, (4)5.0, (5) 7.0 and (6)10.0]% v/v

Fig.(5): Polarization curves for mild steel corrosion in 1.0 M H_2SO_4 + 10% EtOH in presence of different concentrations of (a) aqueous extract (b) alcoholic extract of ZSC plant and 0.01 M chloride ions at 30°C.



$[(F)Free, (1)1 \times 10^{-4}, (2)2.5 \times 10^{-3}, (3)5 \times 10^{-3}, (4)1 \times 10^{-2}, (5)2.5 \times 10^{-2} \text{ and } (6)5 \times 10^{-2}]M$

Fig.(6): Nyquist plots for mild steel corrosion in 1.0 M H_2SO_4 +10% EtOH in presence of different concentrations of chloride ions containing 0.5 %v/v of (a) aqueous extract (b) alcoholic extract at 30°



[(F)Free, (1) 1×10^{-4} , (2) 2.5×10^{-3} , (3) 5×10^{-3} , (4) 1×10^{-2} , (5) 2.5×10^{-2} and (6) 5×10^{-2}]M

Fig.(7): Polarization curves for mild steel corrosion in 1.0 M H_2SO_4 +10% EtOH in presence of different concentrations of chloride ions containing 0.5 % v v of (a) aqueous extract (b)alcoholic extract at 30°C.