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Inhibition of the Acid Corrosion of Zinc by Cannabis Plant Extract

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Abstract The effect of extract of cannabis plant on the corrosion of zinc in aqueous 0.5M sulphuric acid was investigated by electrochemical impedance spectroscopy (EIS) and potentiodynamic polarization techniques. EIS measurements showed that the dissolution process of zinc occurs under activation control. Potentiodynamic polarization curves indicated that the plant extract behaves as mixed-type inhibitor. The corrosion rates of zinc and the inhibition efficiencies of the extract were calculated. The results obtained show that the extract solution of the plant could serve as an effective inhibitor for the corrosion of zinc in sulphuric acid media. Inhibition was found to increase with increasing concentration of the plant extract. Theoretical fitting of different isotherms, Langmuir, Flory–Huggins, and the kinetic–thermodynamic model were tested to clarify the nature of adsorption. Effect of temperature on the inhibitive action of cannabis extract for the corrosion of zinc in 0.5M sulphuric acid was investigated and the activation parameters of the corrosion process in absence and presence of cannabis extract were calculated.

Keywords Zinc, EIS, Inhibitor, Polarization, Acidic corrosion

1. Introduction

Zinc is known to play a major role in corrosion protection of steel structures. To enhance the corrosion resistance of the zinc layer applied for steel protection [1, 2], conversion films are commonly produced on its surface. Although chromate has been successfully applied for many years, the fact that it is toxic and potentially carcinogenic has led to the development of alternative non-toxic corrosion inhibitors, such as silicate, molybdate, rare earth metal salts and some organic compounds [3-14]. Consequently, attention has been focused on the corrosion inhibiting properties [15-33] of natural products from plants. As natural products, they are a source of non-toxic, eco-friendly, readily available and renewable inhibitors for preventing metal corrosion. Several investigations have been reported using such economic plant extract. Earlier, Barannik and Putivlova [34] showed that the actual inhibitors in the plant extracts are usually alkaloids and other organic nitrogen bases, as well as carbohydrates, proteins and their acid hydrolysis products. The existing data show that most organic inhibitors act by adsorption at the metal/solution interface. The adsorption process depends on the electronic characteristics of the inhibitor, the nature of the surface, the temperature and pressure of reaction, steric effect, multilayer adsorption and a varying degree of surface site activity. The aim of present work is to test extract of cannabis plant as inhibitor for the acidic corrosion of zinc and discuss their inhibition mechanism.

2. Experimental

Electrochemical impedance and polarization measurements were achieved using frequency response analyzer (FRA)/potentiostat supplied from prostate instrument. The frequency range for EIS measurements was 0.1×10^4 Hz with applied potential signal amplitude of 10 mV around the rest potential. The data were obtained in a three- electrode mode; platinum sheet and saturated calomel electrodes (SCE) were used as counter and reference electrode. The material used for constructing the working electrode was zinc that had the following chemical composition (% wt) 0.00001% As, 0.001% S, 0.001% P, 0.001% Pb, 0.005% Fe, 99.98% Zn was used for the electrochemical corrosion studies in aqueous solutions. The working electrode was fabricated by cutting and shaping them in cylindrical forms. A long screw fastened to one end of the test cylinder for electrical connection. The Teflon gasket thereby forms a water-tight seal with the specimen electrode that prevents ingress of any electrolyte and thus avoiding crevice effect. The leak-proof assembly exposes only glass, only one side of rod was left uncovered as constant surface area in contact with the solution. The sample was wet hand-polished using different grade emery papers 320, 400, 600, and 800 grit finishes starting with a coarse one and proceeding in steps to the fine grit up to a

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mirror finish, washed thoroughly with double-distilled water and finally dried by absolute ethanol, just before immersion in the solution. Each experiment was carried out with newly polished electrode. Before polarization and EIS measurements, the working electrode was introduced into the test of solution and left for 10 min to attain the open circuit potential (ocp) at which the change of ocp with time is 2 mV/min, i.e., the system had been stabilized. The polarization curve measurements were obtained at scan rate of 20mV/min starting from cathodic potential (E_{corr} -250 mV) going to anodic direction. All the measurements were done at 30.0 ± 0.1°C in solutions open to the atmosphere under unstirred conditions. To test the reliability and reproducibility of the measurements, duplicate experiments were performed in each case of the same conditions.

Optical micrographs will be taken by using Euromex optical microscope, with colour video camera that is connected to a personal computer.

Solution preparation

The test solutions were prepared from analytical grade reagents and distilled water: 98% H_2SO_4 was purchased from Aldrich chemicals. Stock solution of plant extracts was obtained by the flowering tops of plants. A 100 g of dry cannabis plant (the flowering tops of plants), that was obtained by permission from public prosecutor, was minced into very small pieces. The minced plant was boiled with water for 5 minutes to remove chlorophyll and water soluble compounds. The solution was filtered and boiled water was discarded. The process was repeated several times until the final discarded boiled water becomes clear. The minced plant was left to dry in fresh air at room temperature. The dry minced plant was refluxed with 100 ml of petroleum ether. The minced plant solution was then filtered through number 1 Watman filter paper. The extract was evaporated to obtain cannabis residue [35]. This residue was dissolved in 100ml ethanol giving the stock solution of cannabis plant. The concentration of the stock solution was determined by evaporating 10ml and weight the residue [36]. The concentration of the stock solution was expressed in terms of gram per liter. For corrosion measurement in aqueous 0.5 M sulphuric acid solution, the cannabis residue was used to prepare different concentrations of cannabis extract in presence of containing 20% ethyl alcohol.

3. Results and Discussions

3.1. Potentiodynamic Polarization Results

Figure 1 shows the potentiodynamic polarization curves of zinc in 0.5M sulphuric acid, in absence and presence of different concentrations of cannabis extract. As seen from this figure addition of the cannabis extract affects both the cathodic and anodic polarization curves indicating that the cannabis extract could be classified as mixed-type inhibitor. In general, for active metals such as zinc in acid solutions, when dissolved oxygen is present, both hydrogen evolution and oxygen reduction reactions will be possible. However, in view of the fact that, the saturated solubility of oxygen in pure water at 25°C is only about 10^{-3} mol dm^{-3} [37] and decreases slightly with concentration of dissolved salts. In addition, the concentration of H_3O^+ in acid solutions, at pH ≈ 0, is high, and since this ion has a high rate of diffusion, consequently, the contribution of the hydrogen evolution reaction at the cathodic process will overcome the oxygen reduction reaction. Therefore, the corrosion of zinc in acid solution proceeds via two partial reactions [38]:

The partial cathodic reaction involves evolution of hydrogen gas

$$\mathbf{2H^+(aq)+2e^- \rightarrow H_2(g)} \quad (1)$$

The partial anodic reaction involves the oxidation of the metal and formation of soluble Zn^{2+}

$$\mathbf{Zn \rightarrow Zn^{2+} + 2e^-} \quad (2)$$

Since cannabis extract affect the cathodic and anodic process, therefore it could be concluded that the cannabis extract retard the corrosion of zinc by controlling both the hydrogen evolution process and oxidation of Zn^+.

The corrosion current density, (i_{corr}), was calculated from the intersection of anodic and cathodic Tafel lines. The values of the electrochemical polarization parameters, corrosion potential (E_{corr}), corrosion current density, anodic and cathodic Tafel line slope (β_a, and β_c) for different concentrations of cannabis extract are given in table 1. The data revealed that, the corrosion current density that is directly proportional to corrosion rate decreases with increasing the cannabis extract concentration. The values E_{corr} (vs. SCE) for zinc were not affected by increasing the concentration of the extract indicating that cannabis extract could act as pickling inhibitor [39] for zinc metal. The percentage of inhibition efficiency (% P) was calculated from the polarization measurements using the relation:

Table 1. Electrochemical polarization parameters of zinc in 0.5M sulphuric acid containing different cannabis extract concentrations

Conc. (ppm)	$-E_{corr}$ (mV vs. SCE)	B_a	B_c	i_{corr} ($mA.cm^{-2}$)	% P
		($mV.decade^{-1}$)			
0.00	1010	41	161	**1.713**	0
0.50	1012	42	121	**0.361**	79
0.75	1020	70	123	**0.345**	80
1.00	1022	72	123	**0.255**	85
2.00	1915	61	123	**0.226**	87
3.00	1003	44	124	**0.184**	89
4.00	1015	52	112	**0.174**	89
5.00	1022	59	105	**0.166**	90
9.00	1021	52	105	**0.120**	93
10.00	1005	37	114	**0.092**	95

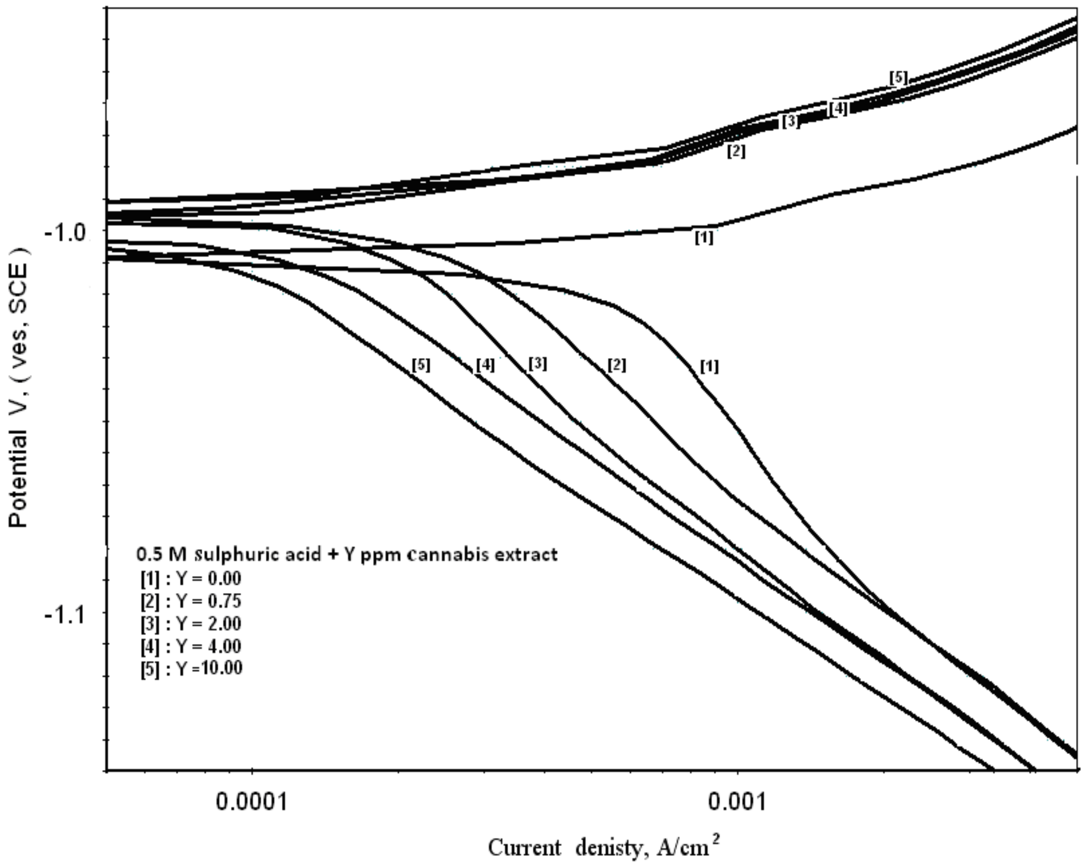

Figure 1. Potentiodynamic polarization curves of zinc in 0.5M sulphuric acid, in absence and presence of different cannabis extract concentrations

$$\%P=[(i_o-i)/i_o]x100 \quad (3)$$

Where $\mathbf{i_o}$ and **i** are the corrosion current density, in absence and presence of cannabis extract.

Figure 2 shows the relation between the percentage inhibition efficiency, and concentration of cannabis extract for zinc in sulphuric acid solution.

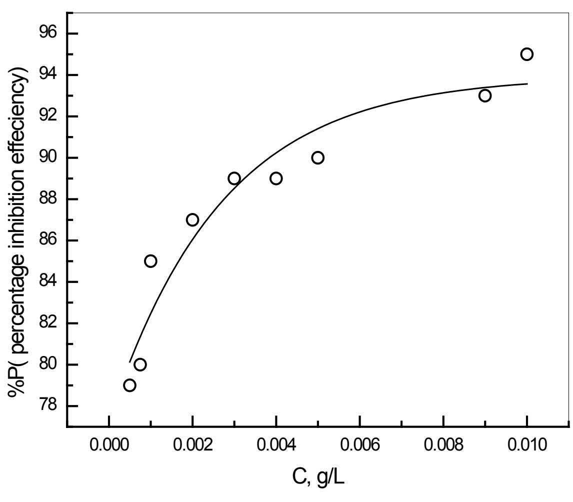

Figure 2. Relation between the percentage inhibition efficiency and concentration of cannabis extract for zinc in 0.5M sulphuric acid solution

The chemical constituents of cannabis extract (cannabidiol-type (CBD) and cannabinol, endogenous cannabinoids anandamide (AEA), 2- arachidonoyl glycerol (2-AG) contain oxygen, nitrogen atoms and π-electrons bonds [40]. Therefore, the adsorption at the metal/solution interface could take place via (i) electrostatic attraction between the charged metal and the charged inhibitor molecules (ii) dipole-type interaction between unshared electron pairs in the inhibitor with the metal, (iii) the π-electrons bonds interaction with the metal, and (iv) a combination of all of the above [41].

3.2. Electrochemical Impedance Spectroscopy (EIS) Results

Figure 3 shows the Nyquist impedance plots of zinc in 0.5M sulphuric acid, in absence and presence of different concentrations of cannabis extract. The Nyquist impedance plots of zinc metal shown in this figure explain that the impedance response consists of capacitive semicircle followed by inductive loop indicating that the dissolution process occurs under activation control. The inductive loop is generally attributed to the adsorption of the species resulting from metal dissolution and hydrogen [42]. As seen, the size of the semicircle, increases with increasing cannabis extract concentration.

The impedance spectra of different Nyquist plots for zinc in 0.5 M sulphuric acid, in absence and presence of different concentrations of cannabis extract were analyzed by fitting the experimental data to the equivalent circuit model shown in figure 4. In this circuit R_s represents the solution resistance; R_p is the polarization resistance associated with the layer of products formed during immersion. R_{ct} is the charge transfer resistance and CPE is constant phase element related to the double-layer capacitance.

It is noted that, the capacitances were implemented as constant phase element (CPE) during analysis of the impedance plots. Two values, Q and n define the CPE. The impedance, Z, of CPE is presented by

$$\mathbf{Z_{CPE} = Q^{-1}\ (i\omega)^{-n}} \qquad (4)$$

Where, $i = (-1)^{1/2}$, ω is frequency in rad s^{-1}, $\omega = 2\pi f$ and f is the frequency in Hz. If n equals one, then equation 4 is identical to that of a capacitor, $Z_C = (i\omega C)^{-1}$ where C is ideal capacitance. For non-homogeneous system, n values ranges 0.9-1 [43]. The L element is related to the inductive effect to represent the inductive loop that appears for zinc in sulphuric acid.

Increasing R_{ct} values with the concentration of the extract, for zinc metal under study, suggesting decrease of the corrosion rate since the R_{ct} value, is a measure of electron transfer across the surface, and inversely proportional to the corrosion rate. The decrease in the Q_{dl} values could be attributed to the adsorption of the chemical constituents of the cannabis extract at the metal surface [44]. The data shown in table 2 indicate that the increase in the extract concentration leads to increase of the charge transfer resistance which is associated with a decrease in the none-ideal double layer capacitance. The percentage of inhibition efficiency (% P) was calculated from the impedance measurements using the relation:

$$\mathbf{\%\ P = [(R_{ct} - R_{cto}) / R_{ct}]\ x\ 100} \qquad (5)$$

Where $\mathbf{R_{ct}}$ and $\mathbf{R_{cto}}$ are the charge transfer resistance, in presence and absence of cannabis extract respectively. The values of %P are in a fair agreement with that obtained from polarization measurements.

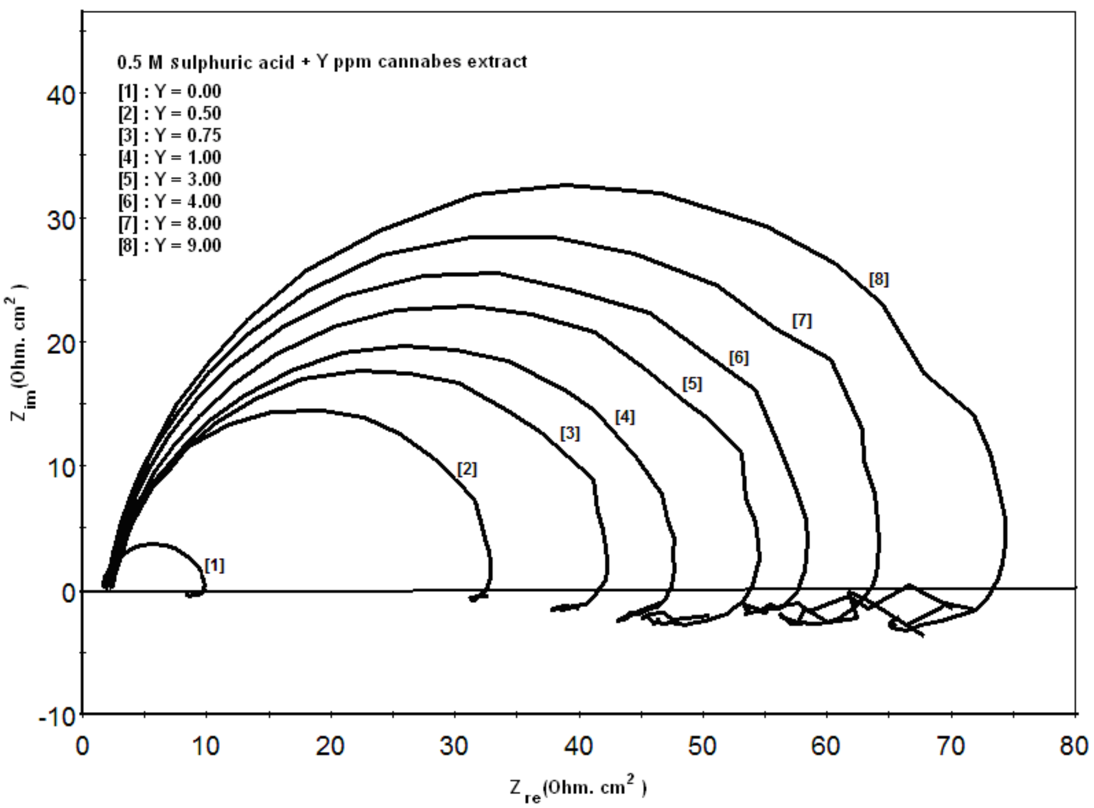

Figure 3. Nyquist plots of zinc in 0.5M sulphuric acid, in absence and presence of different concentrations of cannabis extract

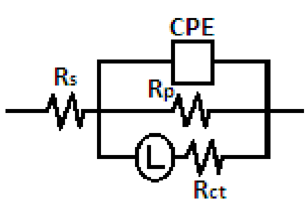

Figure 4. Schematic for the equivalent circuit of zinc

Table 2. Electrochemical impedance parameters of zinc in 0.5M sulphuric acid containing different cannabis extract concentrations

Conc, (ppm)	R_s ($Ohm.cm^2$)	Q_{dl} ($\mu F.cm^{-1}$)	n	R_{ct} ($Ohm.cm^2$)	L	R ($Ohm.cm^2$)	%P
0.00	1.6	57	0.9	**8.1**	18.1	43	0.0
0.50	1.9	54	0.9	**32.3**	0.27	222	74.9
0.75	2.3	51	0.9	**41.7**	19.1	290	80.6
1.00	2.2	44	0.9	**47.1**	36.5	380	82.8
2.00	2.1	43	0.9	**48.2**	45.8	476	83.2
3.00	2.1	34	0.9	**53.4**	41.5	275	84.8
4.00	2.0	33	0.9	**58.2**	35.7	502	86.1
5.00	1.8	30	0.9	**64.5**	58.8	494	87.4
9.00	1.8	29	0.9	**74.7**	44.3	479	89.2
10.00	1.9	27	0.9	**81.5**	75.8	556	90.1

3.3. Optical Micrograph Results

The optical micrographic photos of the zinc samples were captured using optical microscope with magnification power of 40X. Figure 5 shows optical micrographic photos of zinc in 0.5M sulphuric acid free from or containing cannabis extract after 6 hours immersion. As seen from the micrographic photos, the scratch mark of the emery paper still viewed within the experimental exposure time for zinc that was immersed in the acid solution containing cannabis extract whereas in the medium free from the extract, the scratch marks of zinc disappeared due to severe uniform corrosion.

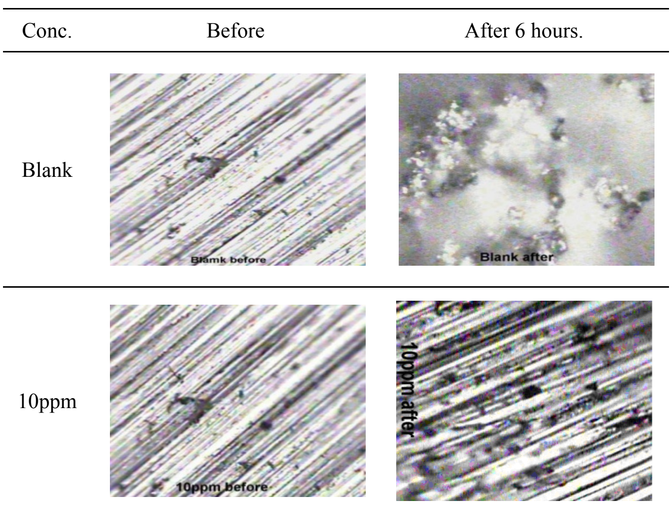

Figure 5. Optical micrographic photos (40X) of zinc in 0.5M sulphuric acid, in absence and presence of cannabis extracts

3.4. Application of Adsorption Isotherms

The understanding of the nature of the adsorption process of various kinds of extracts on metal surfaces was essential to our knowledge of their inhibition action on corrosion. The action of an inhibitor in the presence of aggressive acid media is assumed to be due to its adsorption [45] at the metal/solution interface. The inhibition action was regarded as simple substitutional process [46, 47], in which an inhibitor molecule in the aqueous phase substitutes an x number of water molecules adsorbed on the metal surface, viz.

$$I_{(aq)} + x(H_2O)_{sur} \rightarrow I_{(Sur)} + x(H_2O)_{aq} \quad (6)$$

Where x is the size ratio (the relative size of the inhibitor molecule to the number of surface–adsorbed water molecules) this indicates that the number of adsorbed water molecules displaced depends on the size of the adsorbate.

The degree of surface coverage (θ) of the metal surface by an adsorbed cannabis extract was calculated from polarization measurements using the equations:

$$\theta = (i_o - i)/i_o \quad (7)$$

Langmuir, Flory Huggins isotherms and Kinetic-Thermodynamic model were used to fit the corrosion data of the cannabis extracts.

The Langmuir isotherm is given by [48]

$$[\theta/(1-\theta)]=K[C] \quad (8)$$

Where K is the binding constant representing the interaction of the additives with metal surface and C is the concentration of the additives.

Flory-Huggins isotherm is given by [49]

$$\theta/[x(1-\theta)^x]= K[C] \quad (9)$$

Where x is the size parameter and is a measure of the number of adsorbed water molecules substituted by a given inhibitor molecule.

And the kinetic - thermodynamic model is given by [50]

$$Log[\theta/(1-\theta)]=LogK'+yLogC \quad (10)$$

Where y is the number of inhibitor molecules occupying one active site. The binding constant K is given by:

$$K=K'^{(1/y)} \quad (11)$$

Figures (6-8) show the application of the above mentioned models to the data of cannabis extract obtained from polarization measurements for zinc surface. The parameters obtained from the figures are depicted in table 3.

It is clear that the Langmuir isotherm is unsuitable to fit the data of zinc indicating that there might be non-ideal behavior in the adsorption processes [51] of cannabis extract on zinc surface. On the other hand, Flory-Huggins isotherm and Kinetic-Thermodynamic model are found to be applicable. The values of the size parameter x for zinc indicates that the adsorbed species of cannabis extract is bulky since it could displace more than one water molecule from the zinc surface [52]. The number of active sites occupied by a single inhibitor molecule, 1/y, were nearly equal to the size parameter x.

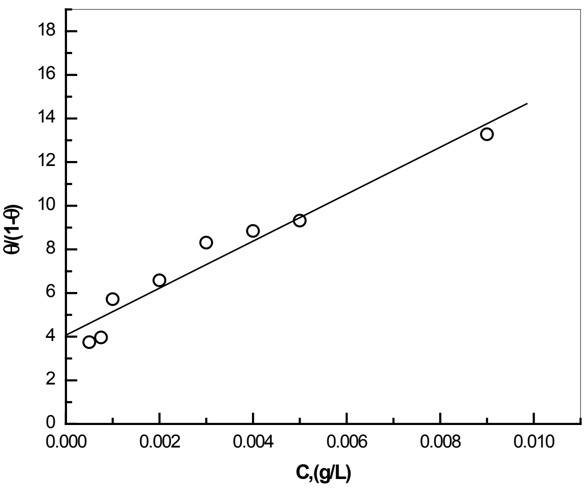

Figure 6. Linear fitting of the data of cannabis extract to Langmuir isotherm for zinc

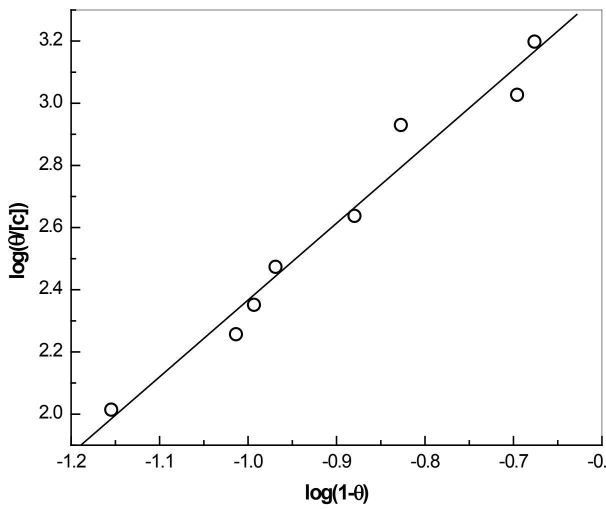

Figure 7. Linear fitting of the data of cannabis extract to Flory Huggins isotherm for zinc

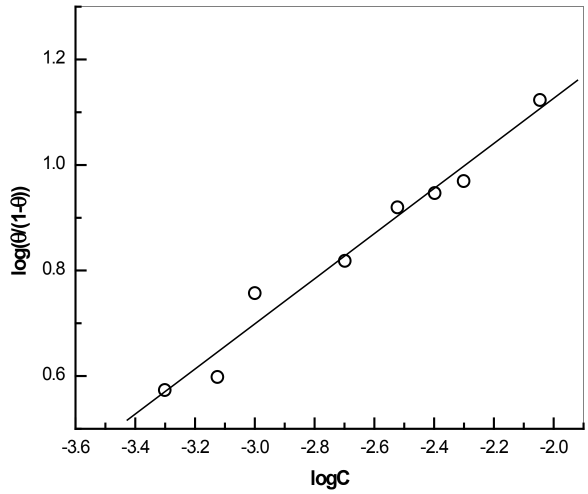

Figure 8. Linear fitting of the data of cannabis extract to Flory Huggins isotherm for zinc

Table 3. Linear fitting parameters of the adsorption of chemical constituents of cannabis on the zinc surface according to the used models

Langmuir	Flory-Huggins		Kinetic-Thermodynamic	
K	K	x	K	1/y
-	17684	2.2	30259	2.1

Since the efficiency of a given inhibitor was essentially a function of the magnitude of its binding constant K, large values of K indicate better and stronger interaction, whereas small values of K mean that the interaction between the inhibitor molecules and the metal is weaker [53]. Hence, according to the numerical values of K obtained from Flory-Huggins or Kinetic-Thermodynamic model, the inhibition efficiency of cannabis extract for zinc higher could be explained on the basis of the mechanism that suggests adsorption of the cannabis extract on the surface of the native metal acting as a film forming species decreasing the active area available for acid attack.

3.5. Effect of Temperature on the Inhibitive Action of Cannabis Extract for the Corrosion of zinc in 0.5M Sulphuric Acid

Many industrial processes take place at high temperatures so, it is particularly important to study the variation of the inhibition efficiency with temperature. When temperature is raised, corrosive action is usually accelerated, particularly in media where evolution of hydrogen accompanies corrosion. Raising the temperature will decrease the inhibitor adsorption on the metal surface; consequently, it will lose its protective action. Cannabis was used in this investigation as an example of plant extract, as a corrosion inhibitor. Figures (9, 10) show the potentiodynamic polarization curves of zinc in 0.5M sulphuric acid, in absence and presence of cannabis extract at different temperatures. It is evident that increasing temperature affects anodic and cathodic parts of the polarization curves but by different degree depending on the solution composition. The values of the electrochemical parameters of zinc in 0.5M sulphuric acid, in absence and presence of cannabis extract at different temperatures are given in table 4. The data show that increasing temperature lead to increase of the corrosion current density (i_{corr}).

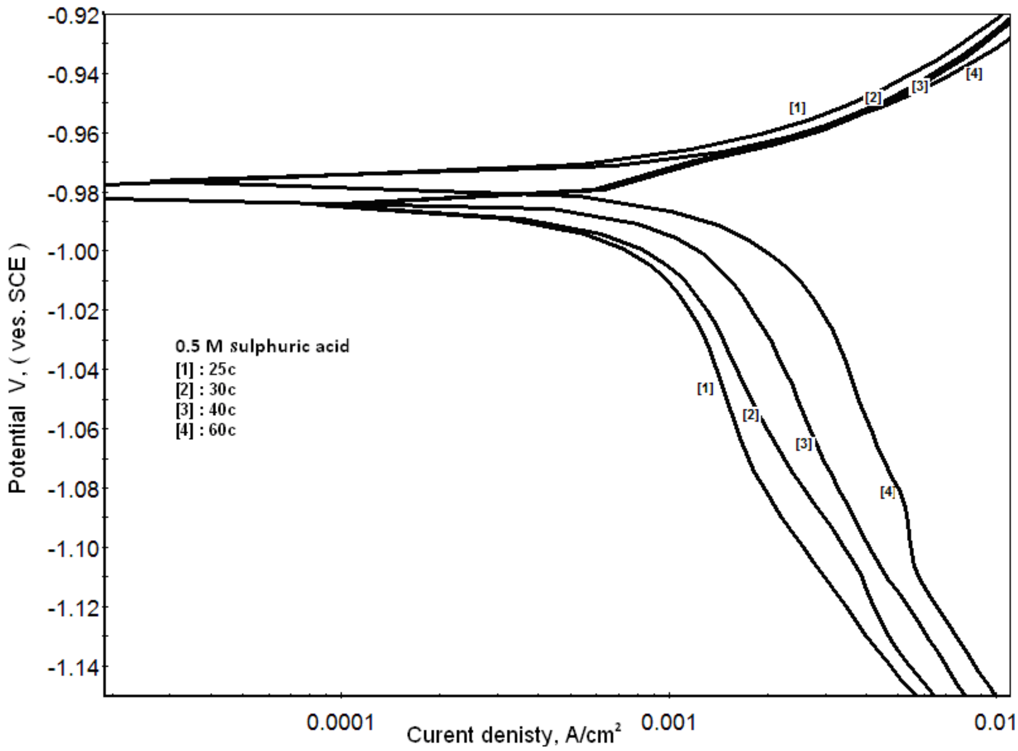

Figure 9. Potentiodynamic polarization curves of zinc in 0.5M sulphuric acid, in at different temperatures

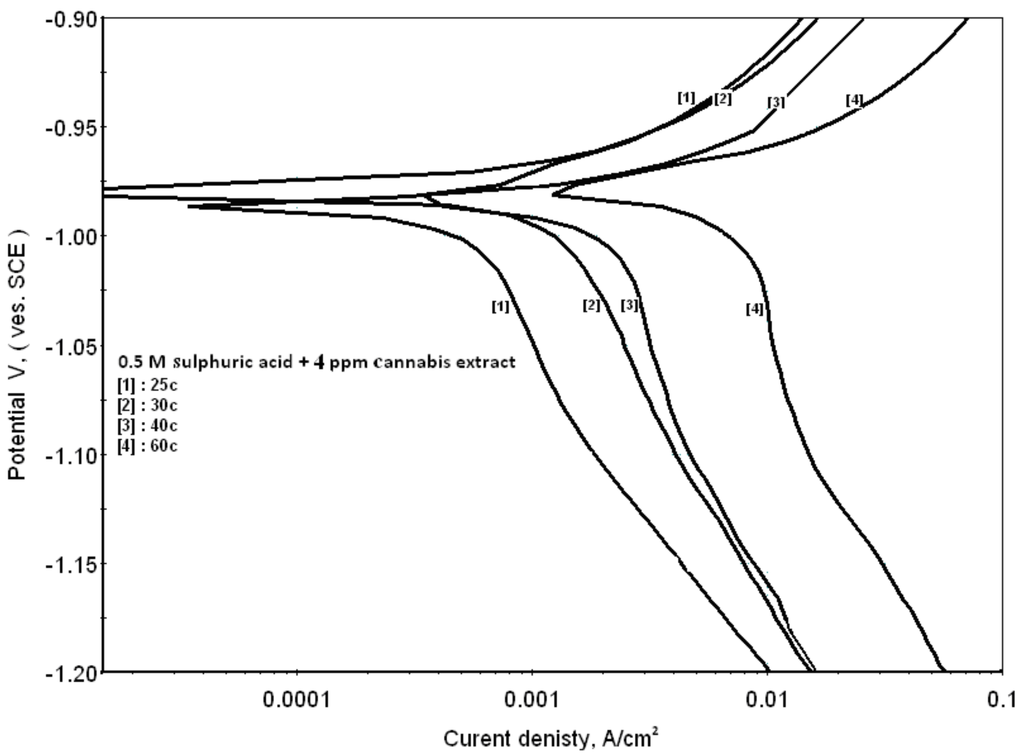

Figure 10. Potentiodynamic polarization curves of zinc in 0.5M sulphuric acid, in presence of 4ppm of cannabis extract at different temperatures

Table 4. Electrochemical polarization parameters of zinc in 0.5M sulphuric acid, in absence and presence of cannabis extract at different temperatures

Conc, (ppm)	Temp. °C	$-E_{corr}$ (mV vs. SCE)	B_a (mV.decade^{-1})	$-B_c$ (mV.decade^{-1})	i_{corr} (mA/cm^2)
Blank	25	992	46	162	**1.676**
	30	997	66	178	**1.948**
	40	999	73	235	**2.727**
	60	1004	85	291	**3.636**
4ppm	25	994	83	147	**1.439**
	30	989	61	180	**1.496**
	40	981	44	275	**1.696**
	60	978	61	260	**2.183**

Determination of the activation parameters

It has been pointed out by many investigators that the logarithm corrosion current density (ln i_{corr}) is a linear function with the reciprocal of the absolute temperature 1/T (Arrhenius equation) [54]:

$$\ln i_{corr}=-E_a/RT+A \quad (12)$$

Where E_a is the apparent effective activation energy, T is the absolute temperature, R is the universal gas constant, and *A* is Arrhenius pre-exponential factor. An alternative formulation of the Arrhenius equation is the thermodynamic formulation of the transition state theory [55]:

$$i_{corr}=(RT/Nh)\exp(\Delta S^*/R)\exp(-\Delta H^*/RT) \quad (13)$$

Where, N is the Avogadro's number, h is the Plank's constant, ΔH^* is the enthalpy of activation, and ΔS^* is the entropy of activation. The activation parameters of zinc in 0.5M sulphuric acid, in absence and presence of cannabis extract were obtained from linear square fit of ln i_{corr} and ln (i_{corr} /T) data *vs.* (1/T) as shown in figures (11, 12).

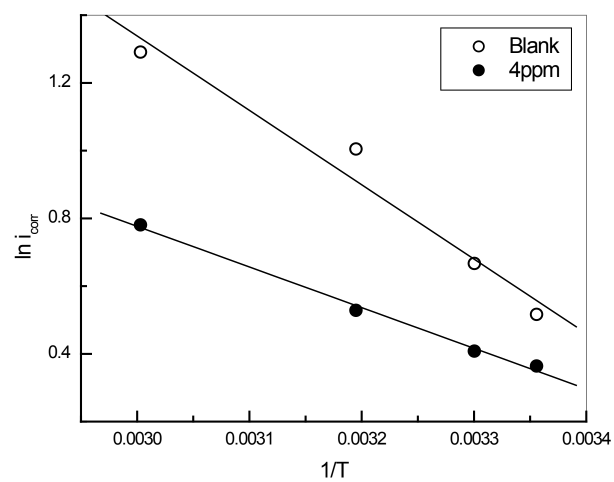

Figure 11. Linear Square fit for zinc of ln i_{corr} *vs.* (1/T)

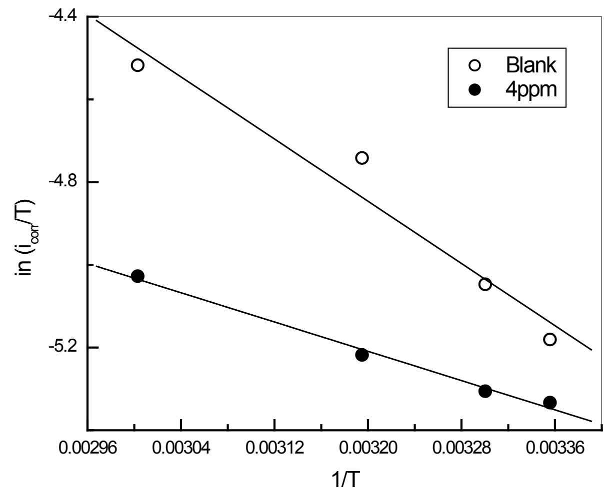

Figure 12. Linear Square fit for zinc of ln (i_{corr}/T) *vs.* (1/T)

The resulting values of the activation parameters are given in table 5. The values of E_a and ΔH^* for dissolution of zinc in sulphuric acid solution containing cannabis extract are lower than in its absence indicates a chemisorption mechanism of adsorption. The value of E_a for zinc in sulphuric acid solution is similar to that obtained previously [56, 57]. The negative value of ΔS^* implies a decrease in the disorder of the system. The decrease in ($-\Delta S^*$) values in presence of the extract may be explained on the basis of increasing the number of chemical constituents extracted from cannabis which are not involved in the adsorption process.

Table 5. Thethermodynamic parameters of activation concerning zinc corrosion in 0.5M sulphuric acid, in absence and presence of different concentrations of cannabis extract

Solution	Activation parameters		
	E_a J/mol.	ΔH^* J/mol.	ΔS^* J/mol.K
0.5M sulphuric acid	18257	15638	-188
0.5M sulphuric acid + 4ppm of cannabis extract	9977	7858	-161

4. Conclusions

1. Cannabis extract act as efficient mixed type inhibitor for the corrosion of zinc in 0.5M sulphuric acid.
2. Inhibition was found to increase with increasing concentration of the cannabis extract.
3. EIS measurements showed that the dissolution process of zinc occurs under activation control.
4. Flory-Hyggins isotherm and Kinitic-Thermodynamic model were found to be applicable to fit the data of adsorption of cannabis at the zinc surface. The data showed that the adsorbed species of cannabis extract are bulky and two adsorbed water molecules were displaced by each species.

5. The values of E_a and ΔH^* for dissolution of zinc in sulphuric acid solution containing cannabis extract are lower than in its absence indicates a chemisorption mechanism of adsorption.

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