

Evaluation of the Ceftriaxone as Corrosion Inhibitor for Carbon Steel Alloy in 0.5M of Hydrochloric Acid.

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Ceftriaxone is drug is evaluated as corrosion inhibitor for carbon steel alloy in presence of 0.5M hydrochloric acid HCl at different concentrations i.e., (10-50) ppm and different temperature. The corrosion inhibition of carbon steel in 0.5M HCl solution by Ceftriaxone was studied at temperature range 293-323K by an electrochemical technique (Tafel method) the results obtained at 50° C showed that this pharmaceutical compound had 93.37% inhibition efficiency at an optimum concentration of 50ppm .the outcomes show that inhibition takes place by adsorption of the inhibitor on metal surface without altering the mechanism of corrosion process. The adsorption of Ceftriaxone takes place according to Langmuir's adsorption Isotherm. Kinetic parameters (activation energy E_a and thermodynamic parameters (enthalpy, entropy and free energy of adsorption ΔH_{ads}° , ΔS_{ads}° , and ΔG_{ads}°) respectively) were calculated and discoursed. An electrochemical method (Tafel method) used to study the behavior of the inhibitors acts as mixed inhibitors.

Keywords: Ceftriaxone, Ofrmax drug, corrosion, carbon steel, Langmuir`s isotherm, acidic media inhibitors.

1. INTRODUCTION

Corrosion inhibition of carbon steel is a matter of theoretical as well as practical importance[1]. Acids are widely used in industries such as pickling, cleaning, descaling, etc., and because of their aggressiveness, inhibitors are used to reduce the rate of dissolution of metals. Compounds containing, S, and O have been reported as inhibitors [2–7]. The efficiency of an organic compound as an inhibitor is mainly dependent upon its ability to get adsorbed on a metal surface, which consists of the replacement of a water molecule at a corroding interface. The adsorption of these compounds is influenced by the electronic structure of inhibiting molecules, the steric factor, aromaticity, electron density at the donor site, molecular area, and molecular weight of the inhibitor molecule [8–11]. In the

present study, the inhibitive properties of the drug Ceftriaxone is reported for the first time, using electrochemical techniques. The inhibition mechanism has been discussed on the basis of these studies.[12]. the effective of an inhibitor must also displace water from the metal surface, interact with anodic or cathodic reaction sites to retard the oxidation and reduction corrosion reaction, and prevent transportation of water and corrosion active species on the surface drug as corrosion inhibitor[13-17]. A few investigations have been reported on the use of antibiotic drugs as corrosion inhibitors[18]. The inhibition action of these drugs was attributed to blocking the surface via formation of insoluble complexes on the metal surface[19-20]. Thus, recently researchers are focusing on Ofrmax(Ceftriaxone) belongs to the group of drugs called cephalosporin antibiotics. It works by fighting bacteria in your body. Ceftriaxone is used to treat many kinds of bacterial infections, including severe or life-threatening forms such as meningitis. Therapeutic actions, Ceftriaxone works by inhibiting the mucopeptide synthesis in the bacterial cell wall[21]. The aim of the new study is evaluated the drugs like Ceftriaxone as corrosion inhibitors for alloy i.e., carbon steel alloy where, Ceftriaxone as drug is non toxic, an environment friendless , inexpensive as well as it has may different hetero atoms like O, S and N atoms which made it as corrosion inhibitor. These properties lead us study a Ceftriaxone as inhibitor rather than synthesis an organic compound that may be toxic, expensive where, Ceftriaxone exhibit a good behavior to do this.

2. EXPERIMENTAL

The structure of the drug that called (Ceftriaxone) that used in this study can be shown below:

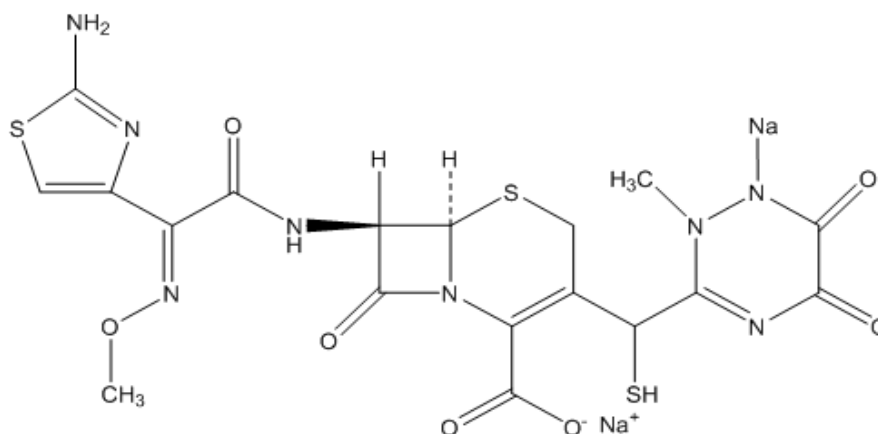


Figure 1. Structure of Ceftriaxone

2.1. Materials

Table 1 represent the composition of the specimens that used to evaluate the Ofrmax drug as corrosion inhibitor for carbon steel alloy as shown below.

Table 1. Composition of carbon steel alloy that used in this study.

C	Mn	Si	P	S	Cr	Fe
0.31	1.10%	0.19	0.04	0.006	0.20	Reminder

2.2. Preparing the specimen

In this study the specimen of carbon steel used as disks of 1.4cm diameters and 2 mm thickness. The opposing faces of each specimen were then grind and polished by emery cloth paper to 600 micron. Thereafter, the specimens were degreased with acetone, washed with distilled water and ethanol and dried with hot air. The specimens were stored in a desiccators containing silica gel during the interval between polishing and polarization measurements. During the test the specimen was placed on the holder metal base and inserted through electrode mounting rod, which was screwed onto the rod by Teflon cover to isolate the disc and to avoid leakage from the working environment. The working electrode was carbon steel of composition that stated above. The exposed area of each ample was 1 cm² and the rest being covered as stated above. A glassy carbon rod was used as counter electrode and saturated calomel electrode (SCE) as reference electrode. The working electrodes were polished following the procedure, described above.

3.RESULTS AND DISCUSSIONS:

3.1. Effect of inhibitor concentration

The corrosion rates of carbon steel in the presence of Ceftriaxone in 0.50 M HCl at different concentrations at constant temperature under study are summarized in Tables 2-5 as bellow:

Table 2. Corrosion data at different concentration for Ceftriaxone at constant temperature 20°C.

Compound	Temp. (°C)	Conc.(ppm)	I_{corr} ($\mu\text{A}\cdot\text{cm}^{-2}$)	CR(mpy)	$-E_{corr}$ (V)	$R_p \times 10^4$ (Ω)	β_a (V. decade)	β_c (V. decade)	Eff. %
Blank	20	18250	59.35	27.30	0.340	0.0191	0.066	0.096
Ceftriaxone	20	10	36.41	16.75	0.354	0.0315	0.067	0.094	38.65
Ceftriaxone	20	20	20.61	9.48	0.366	0.0452	0.084	0.090	62.28
Ceftriaxone	20	30	15.22	6.99	0.355	0.0592	0.082	0.104	74.63
Ceftriaxone	20	40	11.87	5.46	0.352	0.0780	0.082	0.108	80.00
Ceftriaxone	20	50	9.33	4.29	0.352	0.1548	0.088	0.094	84.28

Table 3. Corrosion data at different concentration for Ceftriaxone at constant temperature 30°C .

Compound	Temp. (°C)	Conc.(ppm)	I_{corr} ($\mu A.cm^{-2}$)	CR(mpy)	$-E_{corr}$ (V)	$R_p \times 10^4$ (Ω)	β_a (V. decade)	β_c (V. decade)	Eff. %
Blank	30	18250	112.40	51.90	0.336	0.0183	0.090	0.100	...
Ceftriaxone	30	10	77.82	35.80	0.332	0.2187	0.083	0.111	31.03
Ceftriaxone	30	20	64.84	29.83	0.325	0.2380	0.076	0.107	42.53
Ceftriaxone	30	30	55.58	25.57	0.330	0.2569	0.079	0.116	50.74
Ceftriaxone	30	40	49.06	22.57	0.323	0.2735	0.080	0.107	56.52
Ceftriaxone	30	50	42.58	19.58	0.331	0.3767	0.086	0.107	62.26

Table 4. Corrosion data for different concentration for Ceftriaxone at constant temperature 40°C.

Compound	Temp. (°C)	Conc.(ppm)	I_{corr} ($\mu A.cm^{-2}$)	CR(mpy)	$-E_{corr}$ (V)	$R_p \times 10^4$ (Ω)	β_a (V. decade)	β_c (V. decade)	Eff. %
Blank	40	18250	408.50	188.70	0.333	0.00525	0.104	0.094	...
Ceftriaxone	40	10	239.90	110.35	0.321	0.2100	0.100	0.095	41.52
Ceftriaxone	40	20	152.48	70.14	0.324	0.4542	0.081	0.107	62.83
Ceftriaxone	40	30	115.87	53.31	0.322	0.4861	0.083	0.100	71.75
Ceftriaxone	40	40	87.83	40.40	0.324	0.5256	0.070	0.096	78.59
Ceftriaxone	40	50	73.92	34.00	0.330	0.7045	0.070	0.097	81.98

Table 5. Corrosion data for different concentration for the Ceftriaxone at constant temperature 50°C.

Compound	Temp. (°C)	Conc.(ppm)	I_{corr} ($\mu A.cm^{-2}$)	CR(mpy)	$-E_{corr}$ (V)	$R_p \times 10^4$ (Ω)	β_a (V. decade)	β_c (V. decade)	Eff. %
Blank	50	18250	1239.00	572.30	0.316	0.0022	0.126	0.127
Ceftriaxone	50	10	401.36	184.62	0.311	0.0550	0.107	0.100	67.74
Ceftriaxone	50	20	218.34	100.44	0.325	0.0637	0.113	0.144	82.45
Ceftriaxone	50	30	133.74	61.52	0.309	0.1840	0.163	0.140	89.25
Ceftriaxone	50	40	105.50	48.53	0.297	0.2165	0.135	0.132	91.52
Ceftriaxone	50	50	82.49	37.94	0.279	0.3179	0.153	0.130	93.37

*The concentration of the HCl is 0.50M but,, it written in ppm in Tables to get uniformly units corresponding with the concentration units for the inhibitor.

From Tables 2-5 , it clear that corrosion rates in 0.50 M HCl solution with the addition of Ofmax, decreased as the concentration of inhibitor was changed towards higher side. These results reveal the fact that the adsorption of inhibitor as well as and surface coverage on carbon steel increases as the inhibitor concentration increased, this can be insisted by increasing the resistance polarization values and the inhibition values i.e., decreasing in corrosion rates in presence of the inhibitor Ceftriaxone.

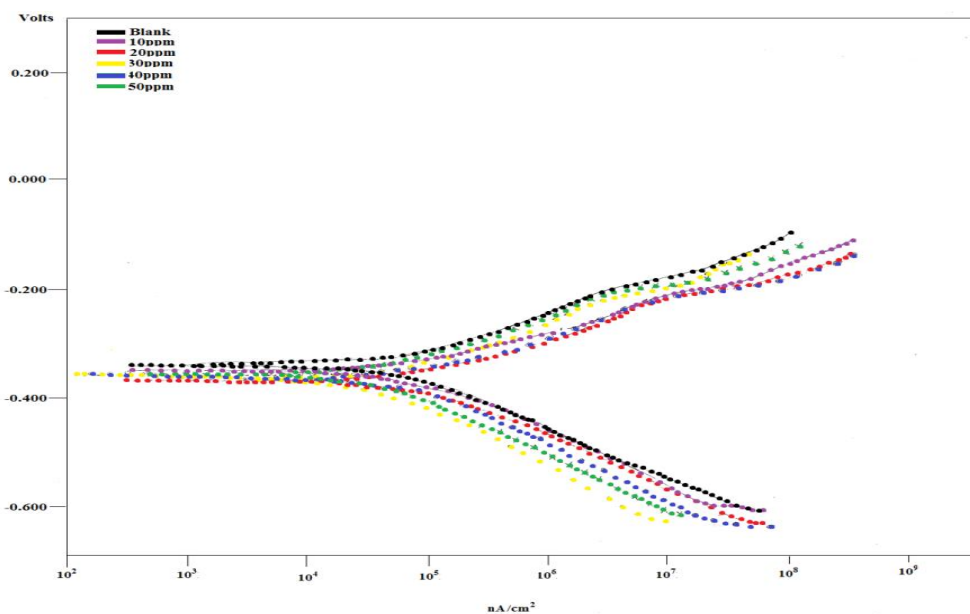


Figure 2. Tafel plot for Ceftriaxone drug at different concentration at 20 °C.

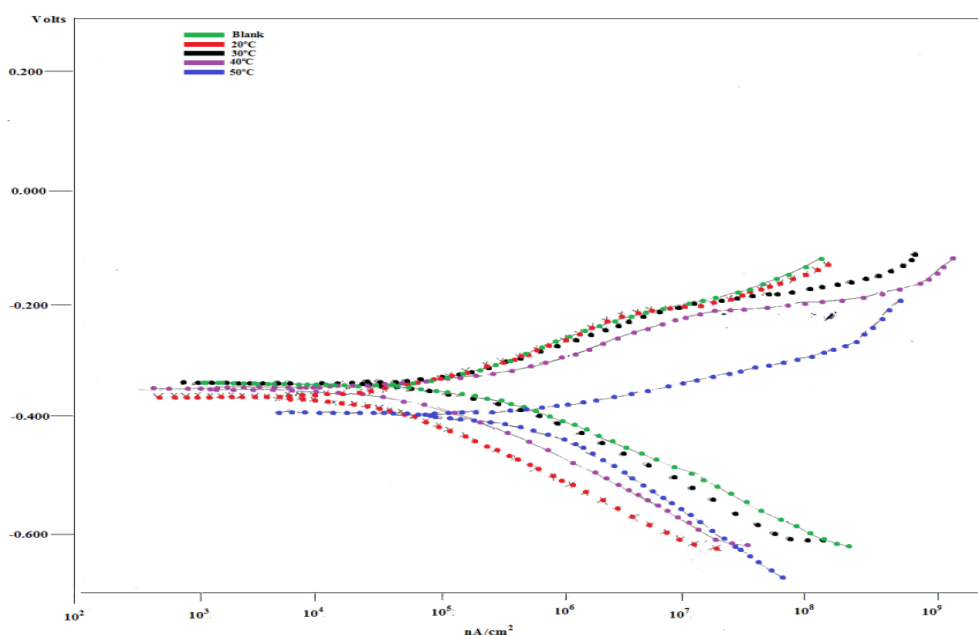


Figure 3. Tafel plot for Ceftriaxone drug at different concentration at 30 °C.

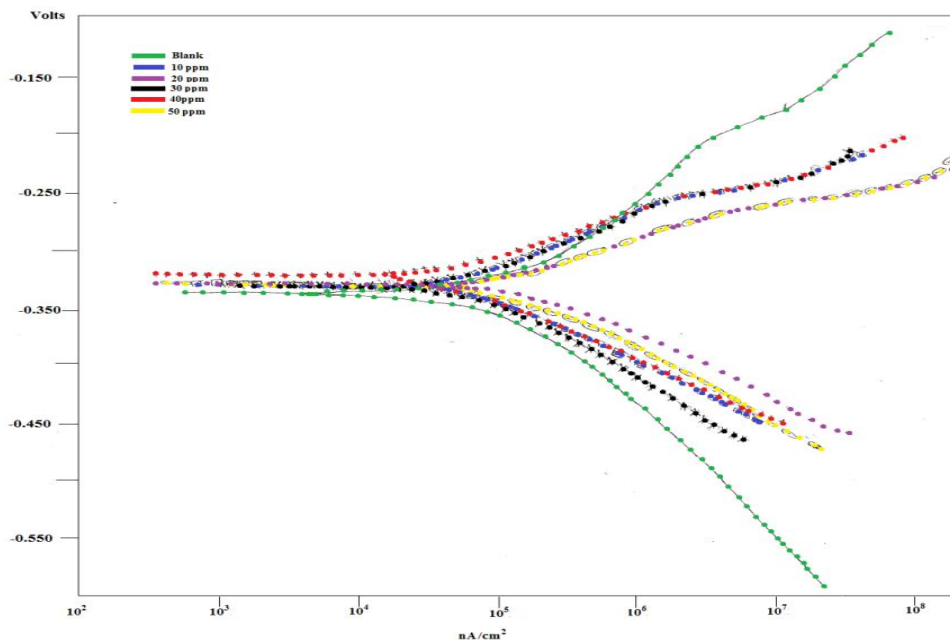


Figure 4. Tafel plot for Ceftriaxone drug at different concentration at 40 °C.

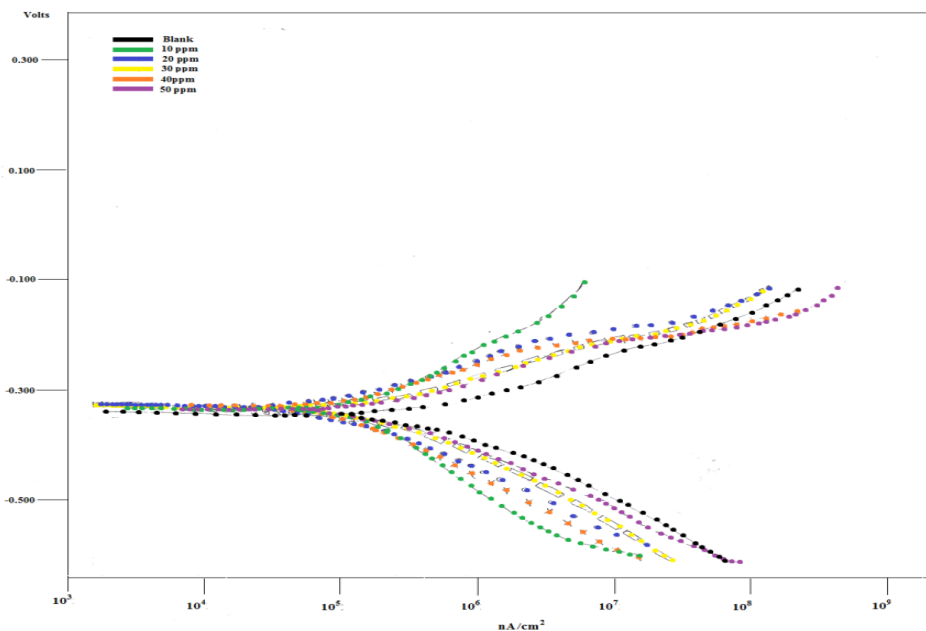


Figure 5. Tafel plot for Ceftriaxone drug at different concentration at 50 °C.

On the other hand, according the above tables anodic behaviors can be observed at temperatures range (30-50) °C while cathodic behavior at 20 °C which meant increasing temperature reduced the hydrogen evolution controlling and the dissolution of the anode (Iron in carbon steel alloy) can be controlled. Although there are disorder values for the β_c and β_a from the above Tables but they

considered as insisting to controlling both the anode dissolution and cathode controlling i.e., the anodic Tafel lines (β_a) are observed to change with addition of inhibitors suggesting that the inhibitor were first adsorbed onto the metal surface and impedes the passage of metal ions from the oxide-free metal surface into the solution, by merely blocking the reaction sites of the metal surface thus affecting the anodic reaction mechanism. Increasing the concentration of the inhibitor gives rise to an inconsistent change in anodic and cathodic current densities indicating that Ceftriaxone acts as a mixed type inhibitor control [22-23]. Tafel plots for Ceftriaxone at constant temperature can be shown in Figure (2-5) as below.

3.2. Effect of temperature:

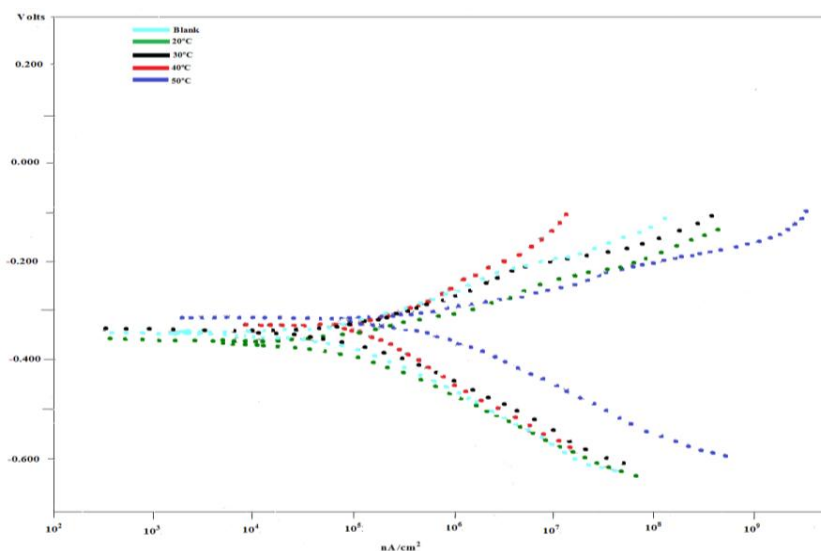


Figure 6. Tafel plot at different temperatures range for 10ppm of Ceftriaxone drug.

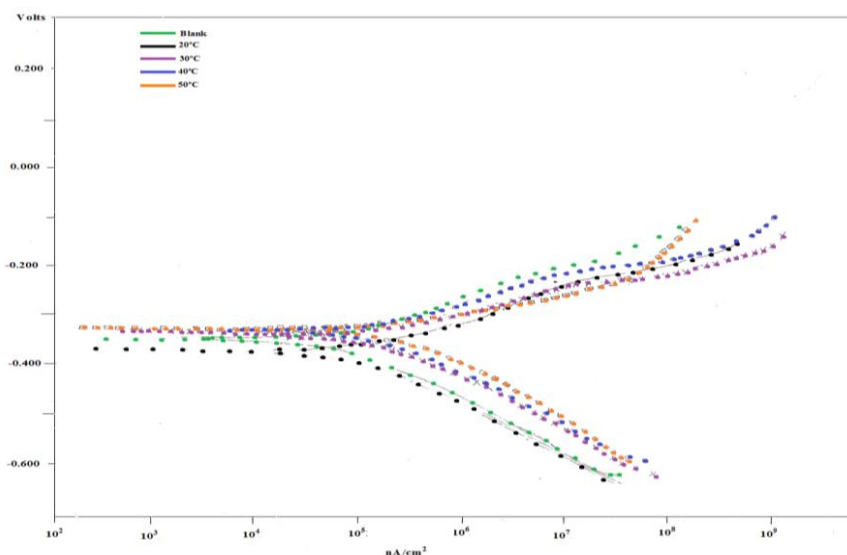


Figure 7. Tafel plot at different temperatures range for 20ppm of Ceftriaxone drug.

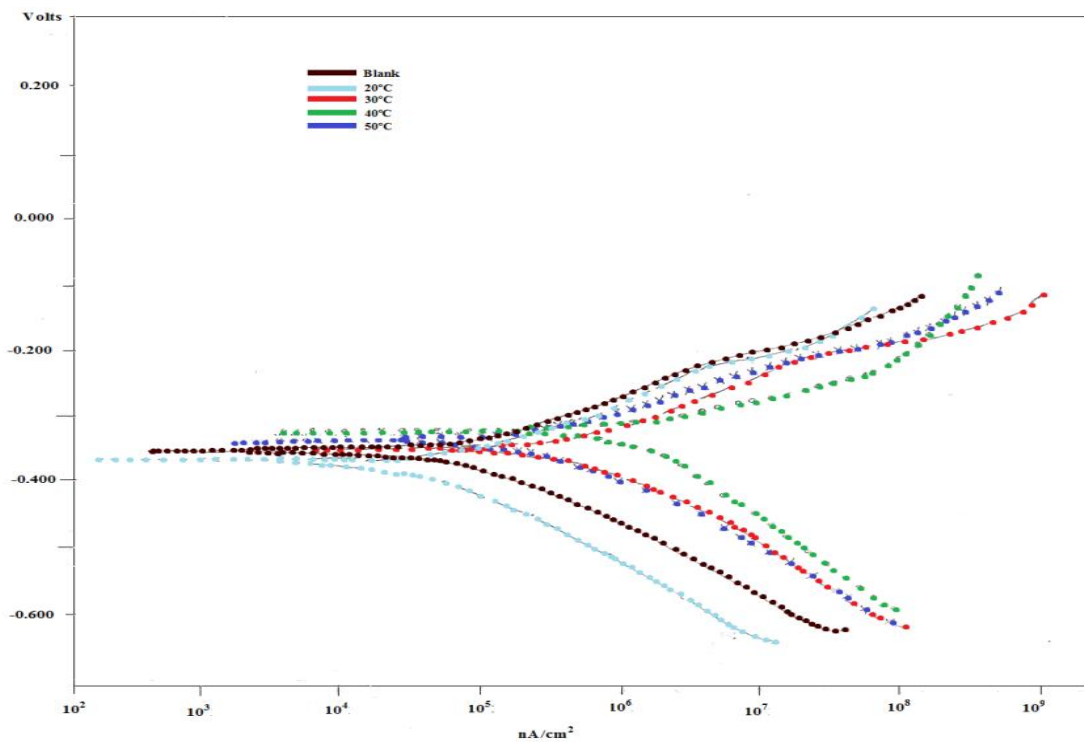


Figure 8. Tafel plot at different temperatures range for 30ppm of Ceftriaxone drug.

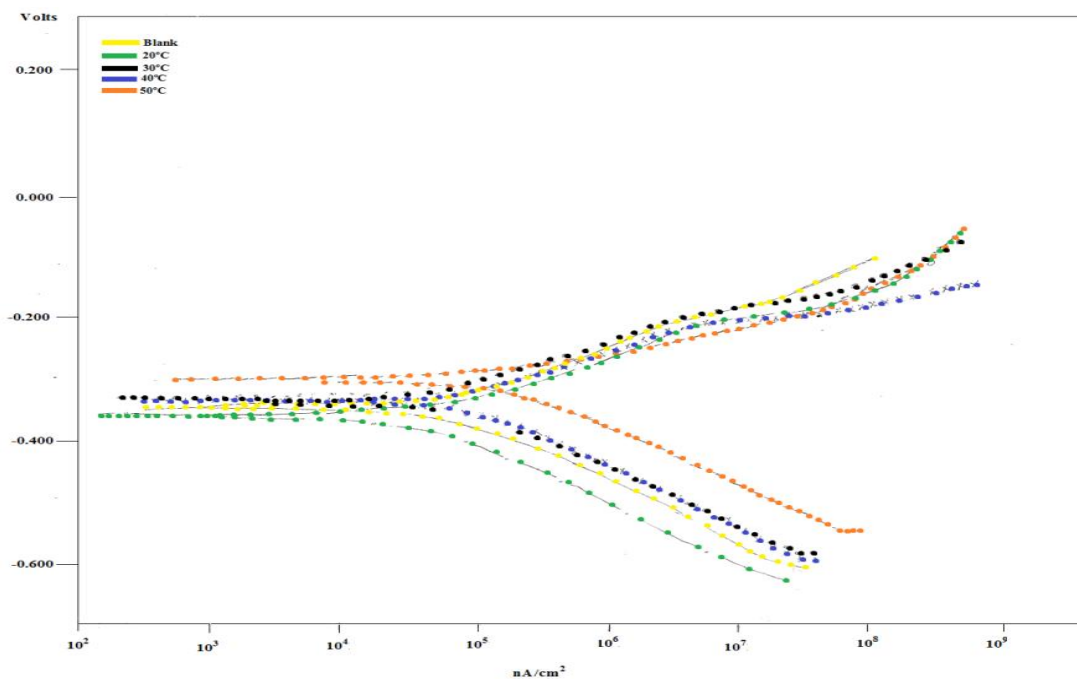


Figure 9. Tafel plot at different temperatures range for 40ppm of Ceftriaxone drug.

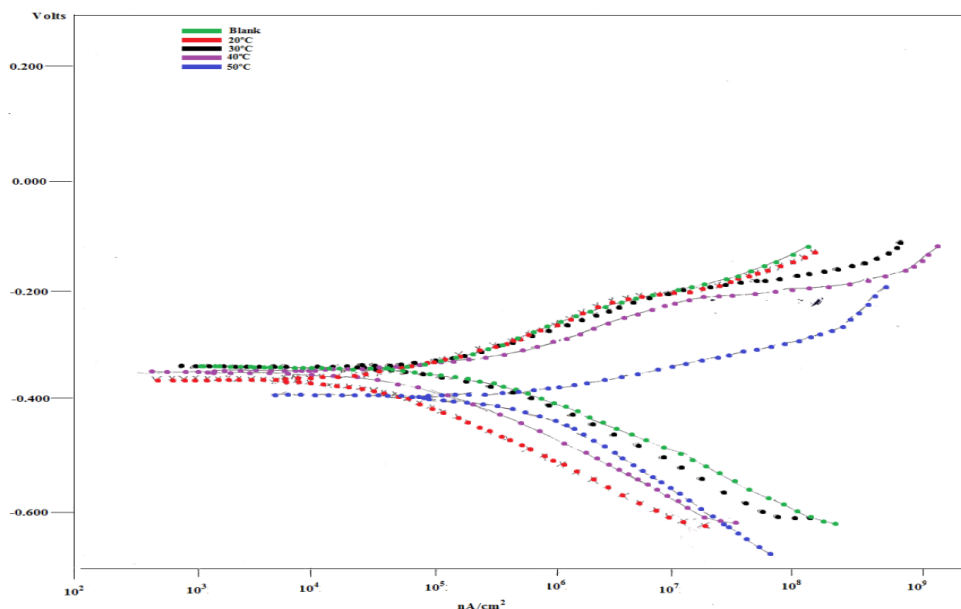


Figure 10. Tafel plot at different temperatures range for 50ppm of Ceftriaxone drug.

On the other hand the data that obtained from the above plots can be tabled in Tables (6-11) as below:

Table 6. Corrosion data for 0.50M HCl(18250*ppm Blank) at different temperatures.

Compound	Temp. (°C)	I_{corr} ($\mu A.cm^{-2}$)	CR(mpy)	$-E_{corr}$ (V)	$R_p \times 10^4$ (Ω)	β_a (V. decade)	β_c (V. decade)	Efficiency%
Blank	20	59.35	27.30	0.340	0.0191	0.066	0.096	...
Blank	30	112.40	51.90	0.336	0.0183	0.090	0.100	...
Blank	40	408.50	188.70	0.333	0.0052	0.104	0.094	...
Blank	50	1239.00	572.30	0.316	0.0022	0.126	0.127	...

Table 7. Corrosion data for 10 ppm of Ceftriaxone at different temperatures.

Compound	Temp. (°C)	I_{corr} ($\mu A.cm^{-2}$)	CR(mpy)	$-E_{corr}$ (V)	$R_p \times 10^4$ (Ω)	β_a (V. decade)	β_c (V. decade)	Efficiency%
Ceftriaxone	20	36.41	16.75	0.354	0.0315	0.067	0.094	38.65
Ceftriaxone	30	77.82	35.80	0.332	0.2187	0.083	0.111	31.03
Ceftriaxone	40	239.90	110.35	0.321	0.2100	0.100	0.095	41.52
Ceftriaxone	50	401.36	184.62	0.311	0.0550	0.107	0.100	67.74

Table 8. Corrosion data for 20 ppm of Ceftriaxone at different temperatures.

Compound	Temp. (°C)	I_{corr} ($\mu A \cdot cm^{-2}$)	CR(mpy)	$-E_{corr}$ (V)	$R_p \times 10^4$ (Ω)	β_a (V. decade)	β_c (V. decade)	Efficiency%
Ceftriaxone	20	20.61	9.48	0.366	0.0452	0.084	0.090	62.28
Ceftriaxone	30	64.84	29.83	0.325	0.2380	0.076	0.107	42.53
Ceftriaxone	40	152.48	70.14	0.324	0.4542	0.081	0.107	62.83
Ceftriaxone	50	218.34	100.44	0.325	0.0637	0.113	0.144	82.45

Table 9. Corrosion data for 30 ppm of Ceftriaxone at different temperatures.

Compound	Temp. (°C)	I_{corr} ($\mu A \cdot cm^{-2}$)	CR(mpy)	$-E_{corr}$ (V)	$R_p \times 10^4$ (Ω)	β_a (V. decade)	β_c (V. decade)	Efficiency%
Ceftriaxone	20	15.22	6.99	0.355	0.0592	0.082	0.104	74.63
Ceftriaxone	30	55.58	25.57	0.330	0.2569	0.079	0.116	50.74
Ceftriaxone	40	115.87	53.31	0.322	0.4861	0.083	0.100	71.75
Ceftriaxone	50	133.74	61.52	0.309	0.1840	0.163	0.140	89.25

Table 10. Corrosion data for 40 ppm of Ceftriaxone at different temperatures.

Compound	Temp. (°C)	I_{corr} ($\mu A \cdot cm^{-2}$)	CR(mpy)	$-E_{corr}$ (V)	$R_p \times 10^4$ (Ω)	β_a (V. decade)	β_c (V. decade)	Efficiency%
Ceftriaxone	20	11.87	5.46	0.352	0.0780	0.082	0.108	80.00
Ceftriaxone	30	49.06	22.57	0.323	0.2735	0.080	0.107	56.52
Ceftriaxone	40	87.83	40.40	0.324	0.5256	0.070	0.096	78.59
Ceftriaxone	50	105.50	48.53	0.297	0.2165	0.135	0.132	91.52

Table 11. Corrosion data for 50 ppm of Ceftriaxone at different temperatures.

Compound	Temp. (°C)	I_{corr} ($\mu A \cdot cm^{-2}$)	CR(mpy)	$-E_{corr}$ (V)	$R_p \times 10^4$ (Ω)	β_a (V. decade)	β_c (V. decade)	Efficiency %
Ceftriaxone	20	9.33	4.29	0.352	0.1548	0.088	0.094	84.28
Ceftriaxone	30	42.58	19.58	0.331	0.3767	0.086	0.107	62.26
Ceftriaxone	40	73.92	34.00	0.330	0.7045	0.070	0.097	81.98
Ceftriaxone	50	82.49	37.94	0.279	0.3179	0.153	0.130	93.37

It shows that inhibition efficiency increased at higher temperatures in case of (10-40) ppm this behavior indicated the chemical adsorption mode[21,24] where the maximum efficiency is 93.37% at 50° C for the concentration 50ppm.

3.3. Adsorption isotherm

The mechanism of corrosion inhibition can be explicated on the basis of adsorption behavior. The degrees of surface coverage (θ) for different inhibitor concentrations in 0.50 M HCl acid in the temperature range (293–323 °K) were assessed by Tafel plot electrochemical method. Data were tested graphically by fitting to various isotherms. By far, the experimental data were best fitted by Langmuir’s adsorption isotherm equation [25]:

$$\frac{\theta}{1-\theta} = K C \dots\dots\dots 1$$

Where C is the concentration of the inhibitor, the isotherms at different temperatures for different concentration of Ofrmax in 0.50M HCl can be explained in Table 12 and Figure 10 respectively:

Table 12. adsorption isotherm data for the different concentration from Ceftriaxone.

compound	Conc.mol/l×10 ⁻⁵	($\theta/1-\theta$) at 293k	($\theta/1-\theta$) at 303k	($\theta/1-\theta$) at 313k	($\theta/1-\theta$) at 323k
Ceftriaxone	1.5	0.63	0.44	0.71	2.10
Ceftriaxone	3.0	1.88	0.74	1.69	4.90
Ceftriaxone	4.5	2.96	1.03	2.63	8.00
Ceftriaxone	6.0	4.15	1.34	3.56	11.00
Ceftriaxone	7.5	5.36	1.65	4.55	14.00

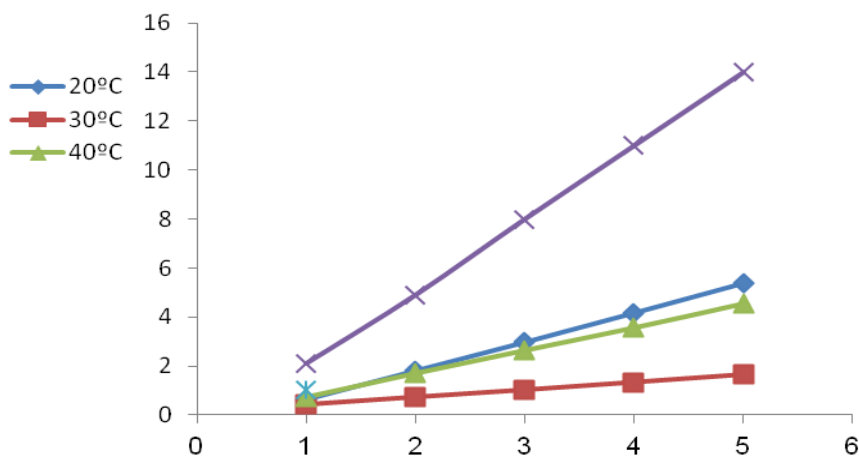


Figure 11. Langmuir’s adsorption isotherm plots for the adsorption of Ceftriaxone at different conc. in 0.50M HCl on Carbon steel surface.

Figure 11 explains the relationship between $\theta/1-\theta$ and C at various temperatures. These results depict that all the slopes are nearly close to unity, which indicates that the adsorption of Ceftriaxone follows Langmuir's adsorption isotherm.

3.4 .Thermodynamic calculations:

Thermodynamic parameters are a very important to study the inhibition mechanism. The thermodynamic functions for dissolution of carbon steel in presence and absence of various concentrations of Ceftriaxone were obtained by applying the Arrhenius equation and the transition state equation [25–27] the data that used to plot as Arrhenius relationship are summarized in Table 13 and the plotting is shown in Figure12:

Table 13. Arrhenius plotting for 0.50M HCl and different concentration from Ceftriaxone.

Temperature K	$1/T \times 10^{-3} \text{ k}^{-1}$	ln(CR) HCl	ln(CR) 10ppm	ln(CR) 20ppm	ln(CR) 30ppm	ln(CR) 40ppm	ln(CR) 50ppm
293	3.413	0.48	3.22	3.27	1.27	2.93	2.65
303	3.300	1.95	3.84	3.60	2.80	3.24	3.39
313	3.201	3.43	4.32	4.19	4.44	3.67	4.07
323	3.096	5.00	5.22	4.61	5.80	3.88	4.30

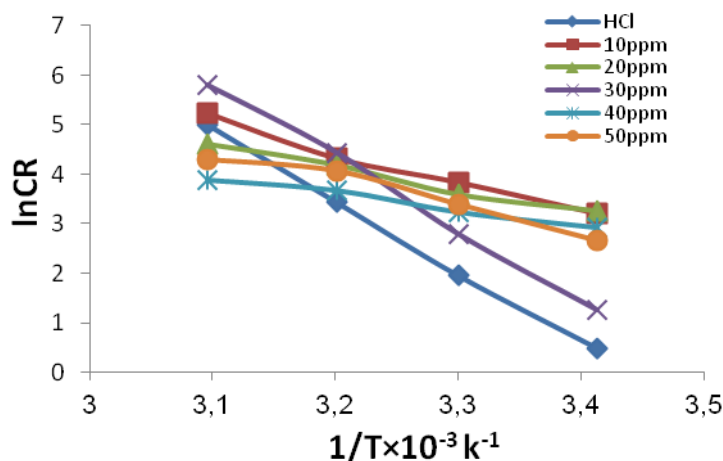


Figure 12. Adsorption isotherm plot for ln (CR) vs. 1/T

The parameters were calculated and presented Table 13 describe that apparent activation energy increased with increasing concentration of Ceftriaxone. This increase in the apparent activation energy may be understood as chemical adsorption [26].

On the other hand, the increase in activation energy can be ascribed to significant increase in the adsorption of the inhibitor molecules on the carbon steel surface with increase in temperature [27]. The parameters were calculated and presented Table 14.

Table 14. Kinetic parameters E^* , ΔH^* , ΔG^* and ΔS^* for carbon steel in 0.50 M HCl without and with addition of different concentrations of Ceftriaxone.

Conc. Of the inhibitor(ppm)	E^* (kJ/mol)	ΔH^* (kJ/mol)	ΔG^* (kJ/mol)	ΔS^* (kJ/mol)
0	5.58	3.14	7.68	0.48
10	8.56	6.12	8.28	5.15
20	9.68	7.24	8.44	5.49
30	10.38	7.94	8.58	6.18
40	11.97	9.53	8.62	5.97
50	12.33	9.89	8.86	7.00

As it is apparent from Table 14, A plot of $\log(CR/T)$ vs. $1/T$ is Straight lines were obtained with slope equal to $\Delta H^*/2.303R$ and intercept equal to $\Delta S^*/2.303R$ from which the values of and were calculated and presented in Table 14 above.

According to Table 14 that the kinetic parameters and of dissolution reaction of carbon steel in 0.5 M HCl in the presence of Ceftriaxone is higher than in the absence of inhibitor. On the other hand, the positive sign of enthalpies refers to an endothermic nature of carbon steel dissolution process i.e. dissolution of steel is difficult [28]. Furthermore, when, the data in the above Table is reviewed, the entropy of activation in mentioned Table clear that these values increased positively in the presence of inhibitor than in its absence. The increase of reveals that an increase in disordering takes place from reactant to the activated complex [29-30].

The standard free energy of adsorption can be calculated according to the relation below:

$$\Delta G_{ads}^\circ = RT \ln(55.5 K_{ads}) \dots\dots\dots 2$$

Where, 55.5 is the concentration of water in solution in mol/l [22]. The values of were calculated from the above equation and summarized in Table 14. Where, R is the molar gas constant and T is absolute temperature. Furthermore, the following equation can be used to calculate a thermodynamic functions:

$$\frac{\Delta G_{ads}^\circ}{T} = \frac{\Delta H_{ads}^\circ}{T} + k \dots\dots\dots 3$$

The variation of $\Delta G_{ads}^\circ /T$ with $1/T$ gives a straight line with a slope that equals ΔH_{ads}° (Figure 13) . It can be viewed from the figure that $\Delta G_{ads}^\circ /T$ decreases with $1/T$ in a linear manner. The calculated values are shown in Table 15. The adsorption heat could be approximately regarded as the standard adsorption heat under experimental conditions. The negative sign of ΔH_{ads}° in HCl solution indicates that the adsorption of inhibitor molecule is an exothermic process[31].

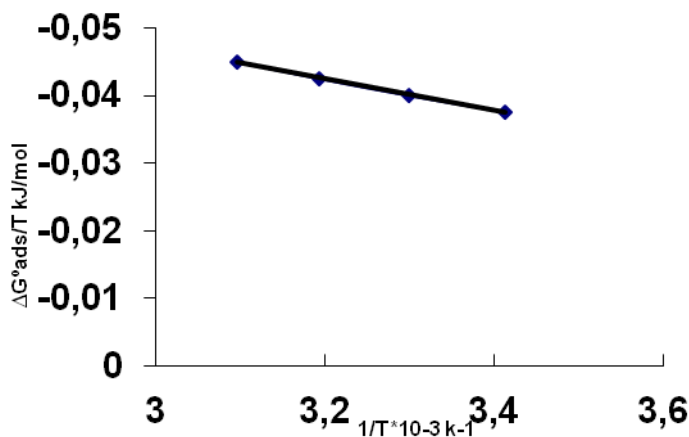


Figure 13. Adsorption isotherm plot for $\Delta G_{ads}^{\circ}/T$ vs. $1/T$.

Then the standard adsorption entropy ΔS_{ads}° was obtained using the thermodynamic basic equation:

$$\Delta G_{ads} = \Delta H_{ads} - T\Delta S_{ads} \dots\dots\dots 4$$

Table 15. Thermodynamic parameters for the adsorption of Ceftriaxone in 1 M HCl on mild steel surface at different temperatures.

Temperature k	ΔG_{ads}° kJ/mol	ΔH_{ads}° kJ/mol	ΔS_{ads}° J/mol
293	-11.03	-23.00	-40.85
303	-11.53	-23.00	-37.85
313	-13.81	-23.00	-29.36
323	-14.31	-23.00	-26.90

The negative values of indicate that the adsorption process is spontaneous and the adsorbed layer on Ceftriaxone surface is stable. Generally, the values of ΔG_{ads}° are negative showed that the adsorption of inhibitor molecules on the metal surface is spontaneous [31-32] . on the other hand, exothermic adsorption reaction data are obtained in addition to ΔS_{ads}° values in the presence of inhibitor are negative, meaning a disordering in presence of inhibitor is reduced in order to the inhibitor is adsorbed on the metal surface. On the other hand, when the data in Table 15 is compared with the inhibition efficiency in Tables 7-11 indicate that increasing in efficiency as temperature increased which is attributed to chemical adsorption mode, thus , at high degree of coverage, the diffusion through the surface layer containing the inhibitor and corrosion products become the rate determining step of the metal dissolution process i.e., the adsorption mode for Ceftriaxone is chemisorptions mode[33].

5. CONCLUSION

Ceftriaxone acts as a good inhibitor for the corrosion of carbon steel in 0.5 M HCl. The inhibition efficiency of Ceftriaxone increased with temperature, which leads to an increase in activation energy of corrosion process. The adsorption of Ceftriaxone follows Langmuir's adsorption isotherm. The adsorption process is spontaneous and exothermic, accompanied by an increase of entropy. Potentiodynamic polarization curves reveals that Ceftriaxone is a mixed-type but.

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