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## Study the Inhibition Effect of Amoxicillin Drug for Corrosion of Carbon Steel in Saline Media

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### Abstract:

Potentiostatic polarization and weight loss methods have been used to investigate the corrosion behavior of carbon steel in sodium chloride solution at different concentrations (0.1, 0.4 and 0.6) M under the influence of temperatures (293, 298, 303, 308 and 313) K. The inhibition efficiency of the amoxicillin drug on carbon steel in 0.6 M NaCl has also been studied based on concentration and temperature. The corrosion rate showed that all salt concentrations (NaCl solution) resulted in corrosion of carbon steel in varying ratio and 0.6 M of salt solution was the highest rate (50.46 g/m<sup>2</sup>.d). The results also indicate that the rate of corrosion increases at a temperature of 313 K. Potentiodynamic polarization studies showed that the examined inhibitor suppress both anodic and cathodic process and behave as mixed type inhibitor. The adsorption of amoxicillin was found to obey Langmuir isotherm model. Arrhenius equation and transition state theory were used to calculate kinetic and thermodynamic parameter. Results obtained showed that corrosion reaction of carbon steel in NaCl is spontaneous and there is a good agreement between the data got from the both techniques employed. SEM analysis was performed to study the film persistency of the inhibitor.

**Key words:** Carbon steel, Kinetic, Sodium Chloride, Thermodynamic.

### Introduction:

Corrosion is a change naturally happening for metals and alloys. The corrosion mechanism of metals and alloys can be explained as follows: when the metal is immersed in the corrosive medium, it begins to oxidize, forming ions inside the solution<sup>1, 2, 1,2</sup>. Studying the corrosion process has a significant attention through preventing and overcoming of this spontaneous process. The corrosion mechanism of different materials are generally depends on the nature of corrosive environment. The existence of the electrolytes in the corrosive media leads to an effect on the rate of corrosion. Sodium chloride is considered as an effective corrosive electrolyte<sup>3, 3</sup>. Steel alloy is extensively used in industry especially for tools and metallic equipment due to its good mechanical properties and low cost. Corrosion is a serious problem for the application of iron and its alloys in many types of service<sup>1</sup>. On the other hand, corrosion process being surface reactions and by

studying surface chemistry, it became certain that surface reactions were directly affected by the presence of foreign particles. For this reason, corrosion should be controlled and the most efficient way is by using compounds known as inhibitors. Inhibitors are mostly organic compounds having heteroatoms like oxygen, nitrogen and sulfur atoms that, when added in small amounts stops or slows down corrosion of metals and alloys. Newly, researchers trend on the use of eco-friendly, cheap and non – toxic inhibitors. Inhibitors are absorbed on the surface of the reactive metal<sup>3,4</sup>. The term adsorption refers to molecules directly linked to the surface, usually only one molecular layer is thick, and does not penetrate into the extent of the metal itself. A known method of controlling corrosion in many branches of technology is the technique of adding inhibitors to the mineral environment. Generally inhibitors adsorb on the entire metal surface impede corrosion reaction<sup>5,6</sup>.

Antibacterial drugs seem to be ideal candidate to replace traditional toxic organic inhibitors due to their natural origin, containing heteroatoms (S, N and O) as active centers, non-hazardous, biodegradable, as well as they could be easily produced and purified. Thus, such investigations are found to be very fruitful and encouraging in saving both metals and environment<sup>7</sup>. Amoxicillin is an antibiotic drug molecule generally having an N-S hetero-cyclic compound containing five oxygen atoms, three nitrogen atoms and one sulfur atom<sup>8</sup>. The molecule is big enough and planar to block more surface area through adsorption on the metal surface, these properties enable it to be an effective inhibitor.

In this research, experimental studies were done to examine the corrosion behavior of carbon steel alloy at various sodium chloride concentrations at five different temperatures in the range 293 – 313 K. Consequently, the inhibition effect of amoxicillin drug at various concentrations and temperatures on the corrosion of carbon steel in 0.6 M sodium chloride solution have been tested using weight loss and potentiodynamic polarization techniques. Furthermore, the kind of interactions between amoxicillin drug and carbon steel surface in 0.6 M NaCl solution were investigated from thermodynamic isotherm simulation.

### Material and Methods:

Analar grade sodium chloride salt was used to prepare the corrosion solutions at various concentrations of 0.1, 0.4 and 0.6 M by dissolving an appropriate amount of NaCl in 1L of deionized water. Amoxicillin (inhibitor) namely, [(2S,5R,6R)-6-[(2R)-2-amino-2-(4-hydroxyphenyl)acetyl]amino]-3,3-dimethyl-7-oxo-4-thia-1-azabicyclo[3.2.0]heptan-2-carboxylic acid], its structure was presented in the Fig. 1 below. Amoxicillin was used as received without any further purification and is added to the 0.6 M NaCl solution at different concentrations of  $6 \times 10^{-6}$ ,  $1 \times 10^{-3}$  and  $6 \times 10^{-3}$  M.

Carbon steel specimen has the following chemical compositions: Carbon (0.1), Silicon (0.24), Manganese (0.47), Chrome (0.12), Molybdenum (0.02), Nickel (0.1), Aluminum (0.03), Cupper (0.14), Cobalt (<0.0012), Vanadium (<0.003), Tungsten (0.06) and the rest is Iron. The specimen with 2.5 cm in diameter and thickness of 1 mm was used as a working electrode in the potentiostatic polarization technique. It was polished by silicon carbide grit abrasive paper from 400, 600, 800, 1200 and 2000, then degreased with acetone and washed with distilled water and finally deride by ethanol. The electrolytic cell had a working capacity of 1L which contained the

working electrode (carbon steel), platinum auxiliary electrode and a saturated calomel reference electrode. The potential-current polarization curves were plotted using a potentiostat (Model WENKING lab-200) obtained from Bank Electronics- Intelligent, GmbH., Germany. This apparatus was commercially programmable, together with an electrometer that provides variability of continuous scan over a desired potential range, including the cathodic and anodic regions.

Loss in weight measurements were performed in 100 mL of sodium chloride solution at concentrations of 0.1, 0.4 and 0.6 molar to immerse three hours of carbon steel at different temperatures in the range of 293 to 313 K. The same experimental procedure was repeated for 0.6 M NaCl solution containing various concentrations of amoxicillin drug of  $6 \times 10^{-4}$ ,  $1 \times 10^{-3}$  and  $6 \times 10^{-3}$  M. These carbon steel specimens were polished as mentioned above, washed with distilled water, degreased with acetone, dried and weighted. After the end of the experiments, carbon steel specimens were washed in distilled water, dried and finally reweighted. Triplet experiments were occurred in each study and the mean weight loss value was reported. From the loss in weight measurements, the corrosion rate ( $C_R$ ) was calculated using the following relation:

$$C_R = (W_1 - W_2) / (S \cdot t) \quad \dots(1)$$

where,  $W_1$  is the weight of steel before corrosion,  $W_2$  is the weight of steel after corrosion,  $S$  is the surface area of the specimen,  $t$  is the immersion time, and  $C_R$  is the corrosion rate. The inhibition efficiency (IE %) was calculated by using the following relation<sup>9</sup>,

$$IE\% = [(C_R^0 - C_R^i) / C_R^0] \times 100 \quad \dots(2)$$

where,  $C_R^0$  is the corrosion rate without inhibitor and  $C_R^i$  is the corrosion rate with inhibitor.

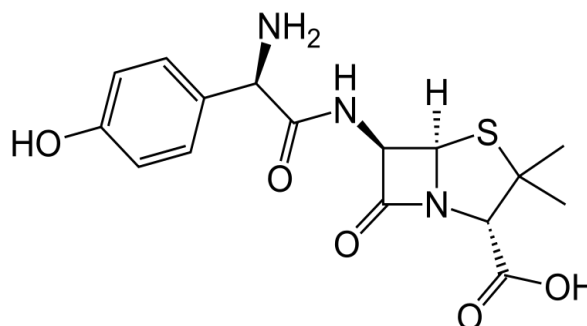


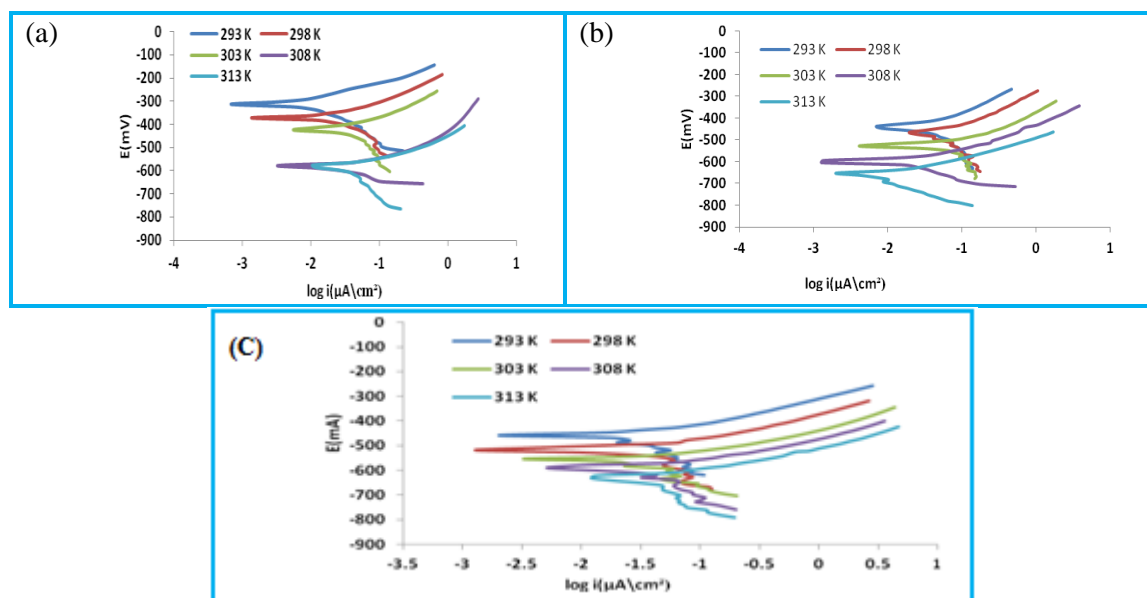
Figure 1. Amoxicillin structure

**Results and Discussion:**

**Potentiostatic Polarization Measurement of Carbon Steel**

In order to notice the electrochemical behavior of carbon steel in various concentrations of sodium chloride solutions and obtain preliminary information about how amoxicillin drug can influence the corrosion process of it, potentiostatic polarization curves were recorded. Fig. 2 a, b, c

represent the typical polarization curves for carbon steel corrosion in various concentrations of NaCl solution from 0.1 to 0.6 M at various temperatures in the range from 293 to 313 K, respectively. The starting potential was -0.4V (SCE) and the scan range extended up to 0.0 V at a voltage scan rate of 10 mV.s<sup>-1</sup>. The data obtained from the polarization curves are listed in Table 1.



**Figure 2.** A carbon steel polarization curve in various concentrations of NaCl solution and temperatures in the range from 293 to 313 K. (a) 0.1 M , (b) 0.4 M and (c) 0.6 M.

**Table 1.** Corrosion parameters of carbon steel in various concentrations of NaCl solution at five temperatures 293, 298, 303, 308 and 313K.

Conc. of NaCl (M)	T (K)	-E <sub>corr</sub> (mV)	i <sub>corr</sub> (μA cm <sup>-2</sup> )	b <sub>c</sub> (mV.decade <sup>-1</sup> )	b <sub>a</sub> (mV.decade <sup>-1</sup> )
0.1	293	341.6	16.58	171.5	116.4
	298	382.2	32.13	133.6	130.1
	303	437.5	43.18	130.9	134.3
	308	579.7	50.10	159.5	165.3
	313	657.2	57.20	101.5	159.9
0.4	293	468.6	31.89	129.9	161.0
	298	485.1	47.56	107.5	140.3
	303	535.5	55.16	131.5	195.9
	308	643.1	67.44	177.2	136.5
	313	704.7	75.91	134.2	103.3
0.6	293	413.6	40.68	177.2	101.5
	298	479.0	58.58	198.0	129.1
	303	562.3	69.95	153.0	132.3
	308	630.4	85.10	160.0	139.1
	313	635.0	98.02	199.0	145.9

Figure 3 a, b and c represent the typical polarization curves for carbon steel in 0.6 M NaCl solution in the presence of various concentrations of amoxicillin drug of 6x10<sup>-6</sup>, 1x10<sup>-3</sup> and 6x10<sup>-3</sup> M at

various temperatures in the range from 293 to 313 K. Table 2 represents the parameters obtained from the polarization curves.

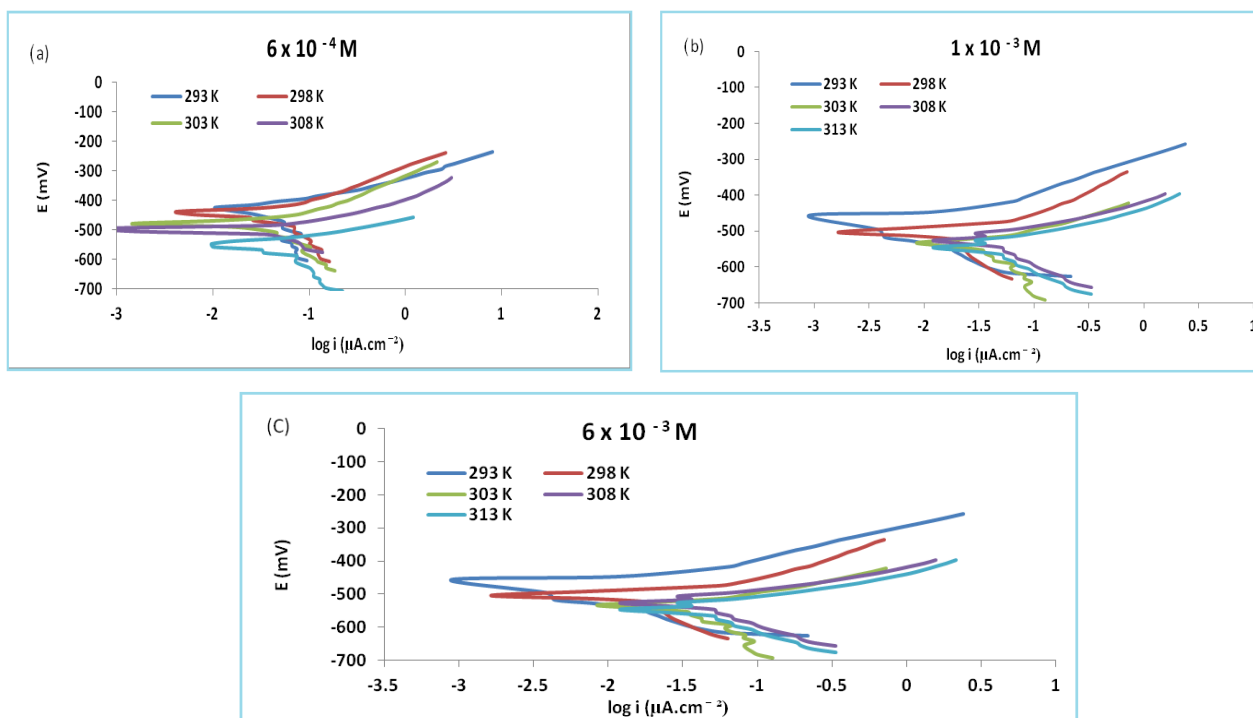


Figure 3. A carbon steel polarization curve in 0.6 M of NaCl solution at various temperatures containing various concentrations of amoxicillin inhibitor.(a)  $6 \times 10^{-4}$  M, (b)  $1 \times 10^{-3}$  M and (c)  $6 \times 10^{-3}$  M.

Table 2. Corrosion parameters of carbon steel in 0.6 M of NaCl solution in the absence and presence of amoxicillin inhibitor at different temperatures in the range 293-318 K.

Conc. of inhibitor (M)	T (K)	$-E_{\text{corr}}$ (mV)	$i_{\text{corr}}$ ( $\mu\text{A cm}^{-2}$ )	$b_c$ ( $\text{mV.decade}^{-1}$ )	$b_a$ ( $\text{mV.decade}^{-1}$ )	IE%	$\theta$
Blank	293	675.2	40.68	377.2	101.5	-	-
	298	699.5	58.58	398.0	129.1	-	-
	303	725.4	69.95	453.0	132.3	-	-
	308	751.3	85.10	460.0	139.1	-	-
	313	779.5	98.02	499.0	145.9	-	-
$6 \times 10^{-4}$	293	518.1	13.37	171.2	143.7	67.13	0.6713
	298	588.2	18.04	164.1	155.6	69.20	0.6920
	303	602.1	19.72	154.0	155.2	71.84	0.7184
	308	625.5	23.65	146.2	151.3	72.23	0.7223
	313	682.5	26.37	137.4	162.1	73.15	0.7315
$1 \times 10^{-3}$	293	413.6	12.93	172.5	112.9	68.22	0.6822
	298	478.9	16.93	166.2	101.8	71.15	0.7115
	303	562.3	19.24	156.3	198.7	72.50	0.7250
	308	630.4	22.47	148.1	147.8	73.61	0.7361
	313	634.9	24.21	140.7	162.5	75.33	0.7533
$6 \times 10^{-3}$	293	388.6	11.92	175.8	123.3	70.66	0.7066
	298	443.2	14.53	168.5	135.5	75.18	0.7518
	303	476.8	16.29	159.3	121.2	76.73	0.7673
		518.2	17.10	150.0	134.0	79.86	0.7986
	313	581.9	17.54	145.5	145.5	82.15	0.8215

It appears from data presented in Tables 1 and 2 that the corrosion potential shifted to more negative values with increasing temperatures in saline solution and to the positive potentials in the presence of various concentrations of inhibitor. The change in the cathodic and anodic Tafel slope

values revealed that the adsorption of amoxicillin drug modify the mechanism of hydrogen evolution reaction in addition to the anodic oxidation. This result indicates that the cathodic and anodic reactions are inhibited and the inhibition efficiency data increase as the concentration of amoxicillin

drug increases<sup>10,11</sup>. On the other hand, the corrosion current density decreases with increasing concentration of inhibitor and increase with increasing temperatures in the range 293-313. Corrosion potential increases with increasing concentration of inhibitor and decreases with increasing temperatures.

The corrosion current densities in the absence and presence of various concentrations of amoxicillin drug in the corrosive medium (0.6 M NaCl) have been used to determine the inhibition efficiency (IE %) using the following equation:

$$IE\% = 100 [ 1 - (i_{corr})_2 / (i_{corr})_1 ] \quad \dots(3)$$

Where  $(i_{corr})_1$  and  $(i_{corr})_2$  are respectively the corrosion current densities of the steel in the absence and the presence of amoxicillin at the same temperature. The results obtained are listed in Table 2. It is indicated that the inhibition efficiency values have been increased with increasing both the amoxicillin concentration and temperature, and these results are similar to data obtained from weight loss method. The surface coverage ( $\theta$ ) was calculated as  $\theta = (1 - (i_{corr})_2 / (i_{corr})_1)$ . Table 2 shows surface coverage values obtained for the corrosion of carbon steel in the presence of various amoxicillin concentrations and various temperatures.

### Polarization Resistance

The overall definition of the polarization resistance of any corroded metal or alloy is the slope of the graphic relationship between the potential (E) and the current density (i) of the corrosion process as follows:

$$R_p = (\partial \eta / \partial i)_{T,C} = [\partial (E - E_c) / \partial i]_{T,C} \quad \dots(4)$$

Where  $\eta$  is the over potential which is defined as the departure of the potential of the electrode from its equilibrium value ( $E_c$ ). for low field approximation ( $\eta \leq 10$  mV) Tafel equation can be miniature to this form

$$\eta = (RT / i_o F)_i \quad \dots(5)$$

Where  $i_o$  is the equilibrium exchange current density.  $R_p$  may best be determined from the equation:

$$R_p = (b_a b_c) / [2.303 (b_a + b_c) i_{corr}] \quad \dots(6)$$

Table 3 shows data resulting from polarization resistance ( $R_p$ ) and equilibrium exchange current ( $i_o$ ) of carbon steel in 0.6 M solution of NaCl containing different concentrations of amoxicillin drug at various temperatures in the range from 293 to 313 K. The  $R_p$  values of carbon steel in 0.6 M NaCl solution decreased with an increase in temperature for uninhibited solution (absence of inhibitor) and increased with increase of both amoxicillin concentration and temperature (Table

3). The increase in polarization resistance indicates that the inhibition efficiency increased with an increase in the concentration of amoxicillin and temperature.

**Table 3. Polarization resistance ( $R_p$ ) and equilibrium exchange current density ( $i_o$ ) values of carbon steel in 0.6 M of NaCl solution in the absence and presence of various concentrations of amoxicillin at five temperatures in the range 293- 313K.**

Additive	T/K	$R_p/10^3$ ( $\Omega/cm^2$ )	$i_o/10^{-4}$ ( $A/cm^2$ )
Blank	293	0.89	0.022
	298	0.82	0.003
	303	0.60	0.012
	308	0.53	0.014
	313	0.44	0.011
$6 \times 10^{-4}$	293	0.98	0.022
	298	0.99	0.024
	303	1.01	0.021
	308	1.07	0.014
	313	1.08	0.015
$1 \times 10^{-3}$	293	1.31	0.073
	298	1.35	0.051
	303	1.48	0.039
	308	1.51	0.016
	313	1.62	0.016
$6 \times 10^{-3}$	293	1.32	0.013
	298	1.41	0.011
	303	1.60	0.009
	308	1.69	0.011
	313	1.76	0.012

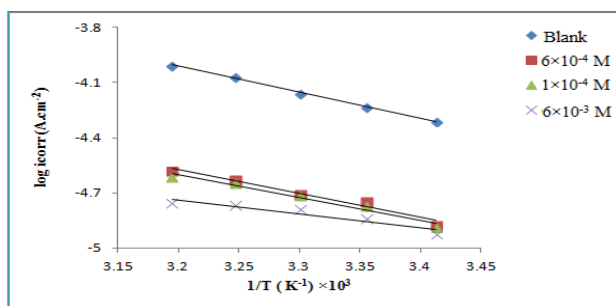
The data in Table 3 show that the polarization resistance decreases as the temperature increases for blank solution and increases with increasing temperature for inhibited solutions. On the other hand,  $R_p$  values are greater in the presence of an inhibitor due to the formation of a protective layer on the surface of carbon steel.

### Kinetics of Corrosion

The rate (r) of carbon steel corrosion at 0.6 M of NaCl solution increased with increasing temperature from 293 to 313K and this behavior followed Arrhenius equation (7),

$$r = A \exp (-E_a/RT) \quad \dots(7)$$

that A and  $E_a$  are respectively, the pre-exponential factor and the energy of activation. Figure 4 shows a plot of  $\log i_{corr}$  values against the reciprocal temperature (1/T) for both the blank (0.6 M NaCl) and presence of various concentrations of amoxicillin inhibitor.



**Figure 4.** Arrhenius plot, relating,  $\log i_{\text{corr}}$  values to  $1/T$  for carbon steel corrosion without and with various amoxicillin drug concentrations at various temperatures in the range from 293 to 313 K.

Table 4 lists the values of Arrhenius parameters ( $E_a$  and  $A$ ) for carbon steel corrosion in 0.6 M NaCl solution in the absence and presence of amoxicillin at different temperatures in the range of 293 to 313K.

**Table 4.** Activation energies ( $E_a$ ) and pre-exponential factors ( $A$ ) data for the corrosion of carbon steel in 0.6 M of NaCl solution without and with various concentrations of amoxicillin over the temperature range from 293 to 313 K.

Additive	T/K	$R^2$	$E_a$ (kJ.mol <sup>-1</sup> )	$A \times 10^{20}$ (molecule.cm <sup>-2</sup> .s <sup>-1</sup> )
Blank	293	0.9739	30.82	4.481
	298			
	303			
	308			
	313			
6×10 <sup>-4</sup>	293	0.9808	46.13	4.646
	298			
	303			
	308			
	313			
1×10 <sup>-3</sup>	293	0.9899	42.28	5.303
	298			
	303			
	308			
	313			
6×10 <sup>-3</sup>	293	0.9599	35.84	6.124
	298			
	303			
	308			
	313			

The data show that the activation energy for steel corrosion in corrosive medium in the presence of inhibitor is higher than those in the absence of it and its value decreases with increasing inhibitor concentration. These results explain why the variations of  $i_{\text{corr}}$  are pronounced in the presence of amoxicillin.

Higher values of activation energy ( $E_a$ ) in the presence of inhibitor refer to a strong inhibitive action by increasing energy barrier for the corrosion process<sup>12</sup>. On the other hand, a decrease of the corrosion activation energy with an increase of inhibitor concentrations, is associated with an increase in inhibition efficiency with increasing temperature as shown in Table 3, referring to the formation of an adsorption layer of chemical nature, which involves the transfer of electrons from amoxicillin molecules to the steel surface, results to the formation of coordinate bond with the surface of carbon steel<sup>13,14</sup>. Alternatively, a chemisorption mechanism associated with increase in inhibition efficiency with temperature and higher activation energy in the presence of the inhibitor<sup>15-17</sup>. In fact, it has been known that chemisorbed molecules are bound at anodic sites of the metal and inhibit the corrosion reaction by retarding the anodic reaction.

### Thermodynamic of Corrosion

The science of energy change, which has been applied for many years to studies corrosion of metals and alloys, is known as thermodynamics. The change in free-energy ( $\Delta G$ ) associated the electrochemical reaction can be calculated using the following equation:

$$\Delta G = - nFE \quad \dots(8)$$

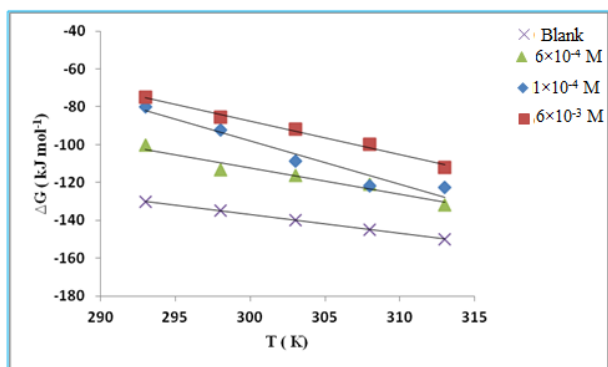
Since the number of electrons in the reaction is denoted by  $n$ , Faraday constant as  $F$  and cell potential ( $E = E_{\text{corr}}$ ) equals  $E$ . Using  $\Delta G$  values obtained from above equation, entropy change ( $\Delta S$ ) of the steel corrosion can be determined using the known thermodynamic relationship:

$$\Delta S = - d(\Delta G) / dT \quad \dots(9)$$

$\Delta G$  data are plotted against temperature and therefor values of  $\Delta S$  can be evaluated from the slope of this plot as shown in Figure 5. At constant temperature, free energy can be expressed as:

$$\Delta G = \Delta H - T\Delta S \quad \dots(10)$$

Where  $\Delta H$  is the change in enthalpy, and  $T$  is absolute temperature. Thermodynamic quantities for the corrosion of carbon steel are given in Table 5.



**Figure 5.** Gibbs free energies ( $\Delta G$ ) data plotted against temperature for the carbon steel corrosion in 0.6 M NaCl solution at without and with different concentrations of amoxicillin.

**Table 5.** Values of thermodynamic quantities for carbon steel corrosion in 0.6 M of NaCl solution without and with various concentrations of amoxicillin inhibitor over the temperature range 293- 313K.

Additive	T/K	$-\Delta G$ (kJ.mol <sup>-1</sup> )	$-\Delta H$ (kJ.mol <sup>-1</sup> )	$\Delta S$ (J.K <sup>-1</sup> .mol <sup>-1</sup> )
Blank	293	130.32	90.18	137.2
	298	135.05	94.22	
	303	140.17	98.67	
	308	145.64	103.44	
	313	150.44	107.56	
6×10 <sup>-4</sup>	293	100.10	58.69	141.0
	298	113.52	70.98	
	303	116.20	73.48	
	308	120.72	76.57	
	313	131.73	86.86	
1×10 <sup>-3</sup>	293	79.82	27.99	176.9
	298	92.44	39.55	
	303	108.52	55.19	
	308	121.66	66.79	
	313	122.55	66.92	
6×10 <sup>-3</sup>	293	75.00	67.58	229.3
	298	85.53	16.76	
	303	92.02	22.61	
	308	100.01	29.47	
	313	112.32	40.64	

Results of Table 5 indicate negative values of  $\Delta G$  that mean, corrosion reactions are occurring spontaneously. The enthalpy changes ( $\Delta H$ ) of the corrosion reaction of carbon steel in different

concentrations of amoxicillin at five temperatures in the range 293-313 K have negative values revealing an exothermic nature for this reaction.  $\Delta S$  values were positive suggesting a lower order of the solvated ions.

### Weight loss study

Corrosion rate data and percentage inhibition efficiency were obtained from loss in weight measurements at different amoxicillin concentrations in 0.6 M NaCl solution after immersion for three hours at different temperatures in the range from 293 to 313 K. The values obtained are listed in Table 6. It has been noticeable that amoxicillin inhibits carbon steel corrosion in 0.6 M NaCl solution at different concentrations used in the research. Table 7 shows that the inhibition efficiency increased from 73.13% to 81.31% with the increasing amoxicillin concentration from 6×10<sup>-4</sup> to 6×10<sup>-3</sup> M at 313 K. On the other hand, corrosion rates of blank saline medium increase with increasing temperature from 293 to 313 K. Furthermore, the corrosion rate of carbon steel in the inhibiting saline medium decreased with increasing temperature. Consequently, the efficiency of amoxicillin inhibition increases significantly with increasing temperature. This finding supports the idea that the adsorption of amoxicillin molecules onto the surface of carbon. Thus, as the temperature increases, the number of adsorbed molecules increases, resulting in increased inhibition efficiency. However, an increased inhibition efficiency (IE%) and a lower corrosion rate ( $C_R$ ) may be due to increased adsorption and increased coverage of amoxicillin molecules on the carbon steel surface with an increased concentration<sup>18</sup>.

**Table 6.** Carbon steel corrosion rates in different NaCl solution concentrations at various temperatures in the range from 293 to 313 K.

	Conc./ M	$C_R$ (g/m <sup>2</sup> d)		
		0.1	0.4	0.6
blank	T (K)			
	293	37.14	39.97	42.12
	298	39.03	41.19	44.40
	303	41.08	43.38	46.69
	308	43.41	45.72	48.13
	313	45.43	47.93	50.46

**Table 7. Corrosion rates ( $C_R$ ) and inhibition efficiencies (IE%) of carbon steel in 0.6 M NaCl, in the presence of different concentrations of amoxicillin at various temperatures in the range from 293 to 313 K.**

Inhibitor	Conc. / M		$6 \times 10^{-4}$			$1 \times 10^{-3}$			$6 \times 10^{-3}$		
	T (K)	$C_R$ (g/m <sup>2</sup> d)	IE%	$\theta$	$C_R$ (g/m <sup>2</sup> d)	IE%	$\theta$	$C_R$ (g/m <sup>2</sup> d)	IE%	$\theta$	
	293	13.95	66.87	0.668	13.39	68.20	0.682	12.36	70.65	0.706	
	298	13.68	69.19	0.692	12.83	71.10	0.711	11.03	75.16	0.752	
	303	13.46	71.82	0.718	12.80	72.49	0.725	10.87	76.72	0.767	
	308	13.37	72.22	0.722	12.71	73.59	0.736	9.69	79.85	0.798	
	313	13.16	73.13	0.731	12.46	75.31	0.753	9.43	81.31	0.813	

### Adsorption isotherms

One of the most important steps in the inhibition mechanism is the adsorption of amoxicillin drug onto the carbon steel surface. Figure 6 shows the  $C/\theta$  data plotted against the concentration of amoxicillin in saline solutions for both potentiostatic polarization and weight loss techniques. The results indicate that the amoxicillin adsorption follows the Langmuir isotherm, which was applied using the relation as follows:

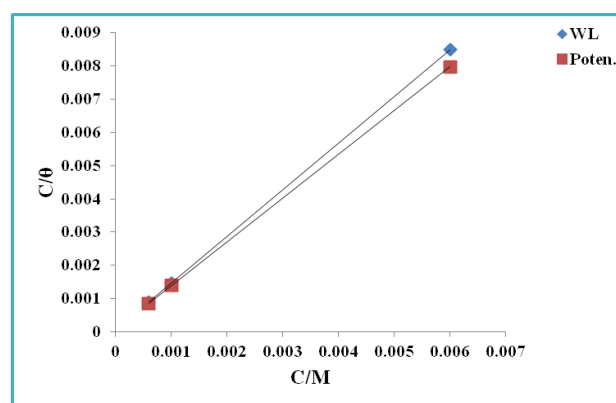
$$C/\theta = 1/K_{ads} + C \quad \dots(11)$$

where  $K_{ads}$  indicates the adsorption constant,  $C$  refers to concentration of amoxicillin, and  $\theta$  represents the surface coverage. Thermodynamic adsorption parameters obtained from the intercept of the straight line obtained by drawing  $C/\theta$  against  $C$  values at various temperatures are listed in Table 8. Table 8 results indicate that the  $R^2$  values (linear regression coefficient) are closed to one, which reveals the amoxicillin adsorption on the surface of carbon steel follow Langmuir adsorption isotherm. Gibbs free energy of adsorption is calculated using the following equation:

$$-\Delta G_{ads} = [\ln K_{ads} - \ln (1/55.5)] RT \quad \dots(12)$$

where  $\Delta G_{ads}$  is the free energy of adsorption, when one water molecule is replaced by one inhibitor molecule and (1/55.5) is the numerical molarity of water. The obtained data are presented in Table 8. The average value of  $\Delta G_{ads}$  was found to be -40.36 kJ/mol. A negative sign indicates that the adsorption of amoxicillin molecules onto the carbon steel surface is spontaneous. It is known that<sup>19</sup> when the value of the free energy of adsorption is equal to -20 kJ / mol and less, the adsorption is physical, whereas when the value is -40 kJ / mol and higher, it indicates that the adsorption is chemical, referring to the charge transfer from the drug molecules to the surface of carbon steel leading to formation of a coordinated type of bond. The obtained value of  $\Delta G_{ads}$  suggests a strong chemical adsorption of amoxicillin molecules onto the surface of carbon steel in saline solution. Values of the

thermodynamic functions of amoxicillin adsorption in saline medium at various temperatures obtained by potentiostatic polarization and weight loss techniques that presented in Table 8. Both methods are of a good agreement.



**Figure 6. Plots of Langmuir isotherm for adsorption of amoxicillin drug on carbon steel surface in 0.6M NaCl solution.**

**Table 8. Thermodynamic parameters for adsorption of the various concentrations of amoxicillin drug on the carbon steel surface in 0.6 M NaCl solution at different temperatures.**

T(K)	Potentiostatic polarization method		Weight loss method	
	$K_{ads}$ (M <sup>-1</sup> )	$-\Delta G_{ads}$ (kJ.mol <sup>-1</sup> )	$K_{ads}$ (M <sup>-1</sup> )	$-\Delta G_{ads}$ (kJ.mol <sup>-1</sup> )
293	179468.7	39.25	145472.1	38.74
298	172858.4	39.83	118861.8	38.90
303	165998.9	40.04	109225.1	39.34
308	163712.0	41.03	109123.3	40.00
313	160873.0	41.64	109043.2	40.65

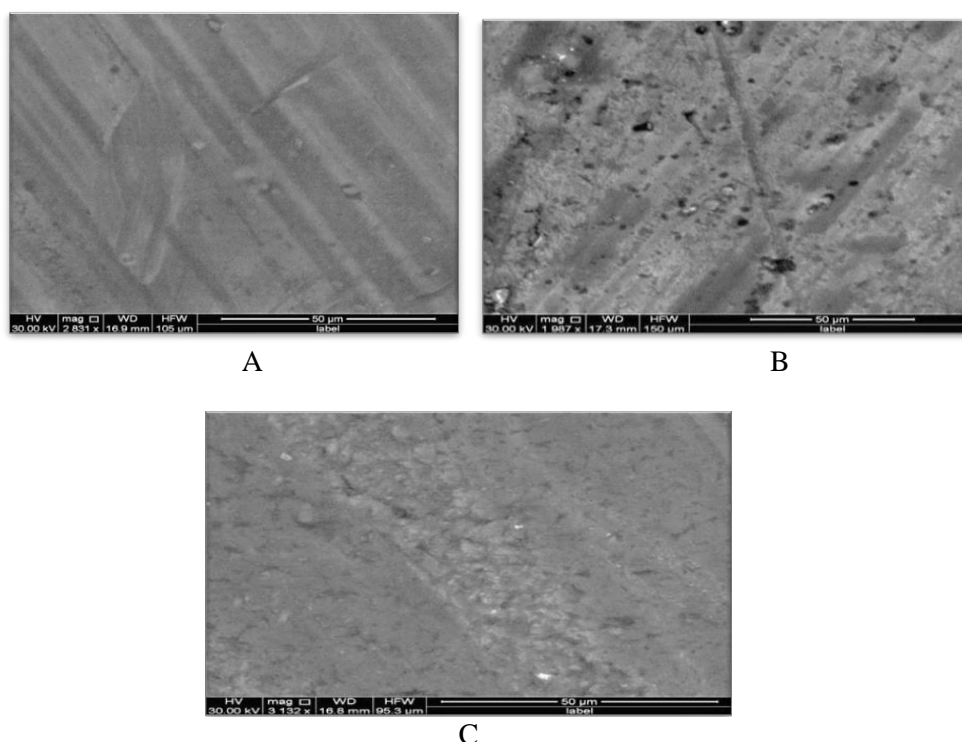
### Scanning electron microscopy

Figure 7 a, b and c. represent scanning electron microscopy of the carbon steel samples. The carbon steel surface before immersion in saline solution seems smooth, as shown in Fig.7a, compared to carbon steel after immersion in uninhibited 0.6 M



NaCl solution for 3 hours. It is clearly shown from Fig.7 b that carbon steel surface shows cracks due to the salt attack on its surface. On the other hand, the presence of  $5 \times 10^{-4}$  M of amoxicillin leads to

reduce the average cracks on steel surface (Fig.7c). Based on what was obtained, it can be concluded that the adsorption layer was efficiently able to inhibit the corrosion of the pure carbon steel sample.



**Figure 7. Scanning electron micrographs of (a) polished carbon steel specimen, (b) in 0.6 M NaCl, (c) in the presence of  $7.5 \times 10^{-4}$  M of amoxicillin.**

### Conclusions:

Based on the results obtained, we can conclude the following:-

- Amoxicillin drug effectively inhibits the carbon steel corrosion in 0.6 M NaCl solution.
- The corrosion process was inhibited by adsorption of the amoxicillin molecules on the carbon steel surface and the adsorption process obey Langmuir adsorption isotherm.
- Both potentiostatic polarization and loss in weight methods indicate that the inhibition efficiency (IE) data increases with increasing amoxicillin concentrations and temperatures.
- Gibbs energy for the adsorption values indicates a general chemical absorption mechanism. This type of inhibitor is effective in room temperature, and is characterized by an increase in the efficiency of inhibition at higher temperatures.
- Polarization curves showed that amoxicillin was a mixed type inhibitor of surface corrosion of carbon steel in 0.6 M NaCl solution.
- The formation of a protective layer on the surface of the carbon steel alloy was confirmed by images obtained by scanning electron microscopy.

### Authors' declaration:

- Conflicts of Interest: None.
- We hereby confirm that all the Figures and Tables in the manuscript are mine ours. Besides, the Figures and images, which are not mine ours, have been given the permission for republication attached with the manuscript.
- Ethical Clearance: The project was approved by the local ethical committee in Al-Nahrain University.

### Authors' contributions statement:

Zeena sh.Mahmoud performed the measurements, were involved in planning and processed the experimental data.

Abeer K. Shams performed the analysis, drafted the manuscript and designed the figures.

Taghried A. Salman aided in interpreting the results and worked on the manuscript. All authors discussed the results and commented on the manuscript

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## دراسة تأثير دواء الأموكسيسيلين لتثبيط تآكل الفولاذ الكربوني في الاوساط المالحة

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### الخلاصة:

تم استخدام طرق الاستقطاب المجهادي الساكن وفقدان الوزن لدراسة سلوك تآكل الفولاذ الكربوني في محلول كلوريد الصوديوم بتركيزات مختلفة (0.1 و 0.4 و 0.6) مولاري تحت تأثير درجات الحرارة (293 و 298 و 303 و 308 و 313) كلفن. كذلك تم دراسة كفاءة تثبيط دواء الأموكسيسيلين على تآكل الفولاذ الكربوني في محلول كلوريد الصوديوم بتركيز 0,6 مولاري على أساس التركيز ودرجة الحرارة. أظهرت النتائج المستحصل عليها ان كل تركيز الملح (محلول كلوريد الصوديوم) أدت إلى تآكل الفولاذ الكربوني بنسب متفاوتة وكان معدل التآكل عند تركيز 0.6 مولاري من محلول الملح هو الاعلى (50.46 جم / م<sup>2</sup> د). تشير النتائج أيضاً إلى أن معدل التآكل لكل تركيز الملح يزداد عند درجة حرارة 313 كلفن. أظهرت دراسات الاستقطاب المجهادي أن المثبط يقلل كل من العمليات الأنودية و الكاثودية ويتصرف كمثبط من النوع المختلط. وجد ان امتزاز الأموكسيسيلين يخضع لنموذج لانكماير متساوي الحرارة. تم استخدام معادلة ارينيوس ونظرية الحالة الانتقالية لحساب المعاملات الحركية والديناميكية الحرارية. أظهرت النتائج التي تم الحصول عليها أن تفاعل تآكل الفولاذ الكربوني في كلوريد الصوديوم هو تلقائي وهناك اتفاق جيد بين النتائج التي تم الحصول عليها من كلتا التقنيتين المستخدمتين. تم إجراء تحليلات بالمجهر الالكتروني الماسح (SEM) لدراسة ثبات الطبقة الواقية للمثبط.

**الكلمات المفتاحية:** الفولاذ الكربوني، حركية، كلوريد الصوديوم، الديناميكية الحرارية.