



## Corrosion Inhibition Property of Amoxicillin Drug for Al-2014 Alloy in 1M HCl Solution

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### Abstract

We focus on the corrosion behavior of Al-2014 alloy in the 1M HCl acidic medium. Also, the inhibition property of Amoxicillin on Al-2014 alloy was studied with different concentrations at different temperatures. The surface analyses through micrographs were analyzed for the inhibitor-treated and untreated Al-2014 surface by FE-SEM measurement. It reveals that the Amoxicillin-treated surface has prevented corrosion compared to the untreated Al-2014 alloy. The potentiodynamic measurements reveal that the Al-2014 alloy with 150ppm of inhibitor solution has the highest tendency to prevent corrosion in 1M HCl solution at different temperatures. The inhibition efficiency is found to be 43% at 25°C and 40% at 35°C. The inhibition efficiency was also verified from the adsorption isotherm, the high inhibition rate is due to the value of ' $K_{ads}$ ' ((6592.3kJ/mole (25°C), 4524.09kJ/mole (35°C)) and  $\Delta G^\circ \approx -33.8$  (25°C), -33.9 (35°C) kJ/mole. From the EIS studies, the inhibition samples show a higher ' $R_{ct}$ ' value implies good corrosion resistance. It is also confirmed from the equivalent circuit by Nyquist plots.

**Keywords:** Al 2014 alloy; Amoxicillin inhibitor; Weight loss; Tafel polarization; Potentiodynamic Polarization; Nyquist plots.

### 1. Introduction

Aluminum is one of the fascinating metals that can find in aerospace engineering because of its unique features in its physical properties and economic affordability. The usage of a material is not just in aerospace but also in the automobile, medicine, etc. [1]. It is also the third most abundant element constituting about 8.13 % of the earth's crust. Its alloys are also the most important materials for aerospace engineering because of their vast electrical and

mechanical properties, such as being lightweight with adequate strength, high conductivity and good corrosion resistance due to the barrier oxide film layer [2]. Aluminum is lightweight; its performance is highly beneficial in transport applications that are related to the aerospace industry [3]. The metal has been widely used as construction materials, heat exchangers, catalysts and power lines. We aim to investigate the corrosion resistance of Al-2014 alloy of the heat-treated [4]. The technological demand is not just fulfilled by their properties at also makes them retain over time. In this work, we are focusing on one of the alloys of Aluminum, i.e. Al-2014 alloy and its corrosion inhibition property by heat treatment [5]. The Al-2014 alloy is the second popular and strongest alloy in the 2000 series, but its usage is limited because of its poor corrosion resistance. Anticipation of metal corrosion is important to increase the lifespan of equipment; normally, it is an important process in aerospace engineering materials. The electronegative heteroatom (N, P, O and S) containing organic compounds act as corrosion inhibitors. Therefore, some of the pharmaceutical antibiotic drug molecules belong to this type of family [6]. The high concentrations of chloride ions may tend to reduce the oxide layer formed against the corrosion [7]. The organic inhibitors 2-mercaptobenzimidazole (2-MBI) tested for the heterogeneous Al-6061 alloys and it act as a protecting layer for corrosion in the acidic medium [8].

The recent studies focused more on the inhibition properties of antibiotic drugs in the various surroundings, it may be an aggressive atmosphere like acidic and alkaline, i.e. Levofloxacin has more electronegative atoms  $\pi$ -bonds, which is the critical factor for a molecule to adhere to the surface of the metals, the inhibition property in the alkaline medium on carbon steel and Aluminum as reported [9]. The gravimetrically estimated Amoxicillin (1800ppm after 8 h immersion) on mild steel in 1 N HCl acts as a mixed inhibitor with an inhibition efficiency of 94.47% [10, 11]. Most recently, Tobramycin acts as an anti-corrosive agent on carbon steel in an acidic medium with inhibition efficiencies of 80 %, 90.5 %, 84.3 % [12]; the antibiotic Azithromycin acts as an inhibitor for corrosion of mild steel, copper and Zinc in a 0.5 M H<sub>2</sub>SO<sub>4</sub> [13]. Also, the corrosion behavior of Aluminum is studied from antibacterial drugs [14], Nifedipine drug [15], the surface morphology was also investigated to observe any change in the micro structure of the heat-treated specimens produced after corrosion immersion test in an aggressive 3.5% NaCl medium. These alloys show specific behavior in Retrogression and Re-aging (RRA) heat-treatments [16]. The most important feature of aluminum alloy is the formation of an oxide layer on the surface which acts as a barrier [17].

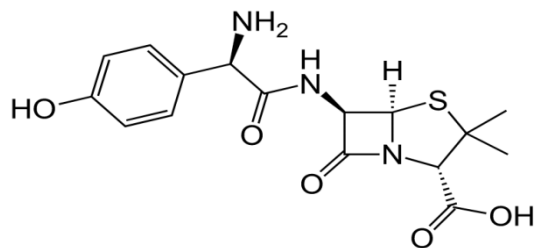
This oxide barrier breaks due to aggressive environments and thus, learning the corrosion parameters of any alloy is essential, to realize and estimate the corrosion current and its response to a metal surface [18].

The previous works reflects inhibitors are in the acidic medium they exhibit the mixed type of adsorption for Aluminium surface and show good inhibition efficiency i.e. Cloxacillin 90 % [19, 20], Clotrimazole 96 % [21], Amine Compound 92 % [22], Chloroquine 94.5 % [23]. The heterocyclic compounds like pyrazole derivatives have a good tendency to act as an inhibitor on mild steel [24] and also several other kinds of research on different layers have been identified. Gece scattered the evaluation article in regards to the preparation of medications utilized as metallic deterioration inhibitors under one in the whole of the clever risky environments [25]. Penicillin subordinates containing  $\beta$ -lactam bundle are typically utilized as antibacterial sellers and appeared for the explanation that the potential open door for usage keenness by methods for the prudence of their make conveying approach, relative low harmful best to circumstance and reasonable atomic structure [26, 27]. Amoxicillin is the one antibiotic drug that is used in the formation of Schiff base molecules. These molecules act as good corrosion inhibitors [28, 29].

## **2. Experimental Techniques**

### **2.1 Materials and Methods:**

The commercially available Al-2014 alloy ingots are selected for the present work and the respective elemental composition is obtained from EDAX spectra. The identified elements are given in table 1. Firstly, Al-2014 was melted by subject the material in a furnace by keeping the temperature slightly just above its melting point. Then, molten compound is poured into preheated cast-iron molds. The specimens were abraded with silicon carbide papers of different grades (from 600-1500) and then polished with the help of a polishing wheel and cleaned using acetone and double-distilled water and then dried at room temperature. The Amoxicillin is bought from Sigma-Aldrich Chemical Co., Inc. and its molecular structure is given in figure 1. A standard 1M HCl solution was prepared in distilled water using Analytical grade salt. The inhibitor (50ppm to 150ppm) is prepared by dissolved in double distilled water and added to a standard 1M HCl solution.



**Figure 1:** The chemical structures of the Amoxicillin Compound

## 2.2 Microstructure and Elemental Analysis

The microstructure and Corrosion morphology of Al 2014 alloy were investigated by scanning electron microscopy (FE-SEM). The work specimen surface is examined after the inhibition and before the inhibition. Similarly, EDAX -measurements are also done in the same instruments after the electrochemical tests were observed by FE-SEM.

## 2.3 Weight loss measurements:

The pre-weighted Al-2014 (2.5 cm × 2 cm × 0.025cm dimension) are immersed in the prepared electrolyte containing 250 ml of 1.0 M HCl with and without inhibitors at 25°C and 35°C for 24 h. Then the samples are pulled back and subjected to a twofold wash with deionized water and (CH<sub>3</sub>)<sub>2</sub>CO. Later, all the samples are dried at room temperature. The weight loss of all the samples is gravimetrically estimated with accuracy (99.8%). The disintegration charge in terms of corrosion rate (C<sub>R</sub>) is estimated from relation (1) [30] and also, the inhibition efficiency is estimated from the following relation (2) [31-34].

$$CR (\text{Corrosion rate (mpy)}) = \frac{534W}{DAT} \quad (\text{mg cm}^{-2}\text{h}^{-1}) \quad (1)$$

Where: W=Weight loss (mg), D=Density of the specimen (g/cm<sup>3</sup>), A=area of the specimen (square Inch), T=Exposure time (hours)

$$(\eta_{WL} \%) = \frac{W_0 - W_i}{W_i} \times 100 \quad (2)$$

Where:  $W_0$  and  $W_i$  are the normal weight reduction of three equal Al 2014 amalgam examples in the absence and presence of inhibitors, individually for 24h drenching.

## 2.4 Electrochemical Analysis

### a) Tafel polarization measurements

Electrochemical analyses are carried out by using CH-Instrument (USA) as per ASTM G3-73 standard. The measurements are carried out by subject-prepared samples of Al-2014 alloy. The working terminal ends up being initially immersed inside the test electrolyte for 30 minutes to accumulate a uniform Open-circuit potential (OCP) - 250 to +250 mV. The polarization rate is measured in terms of a potential rate 0.166mVsec<sup>-1</sup> [35].

**b) Electrochemical Impedance Spectroscopy,**

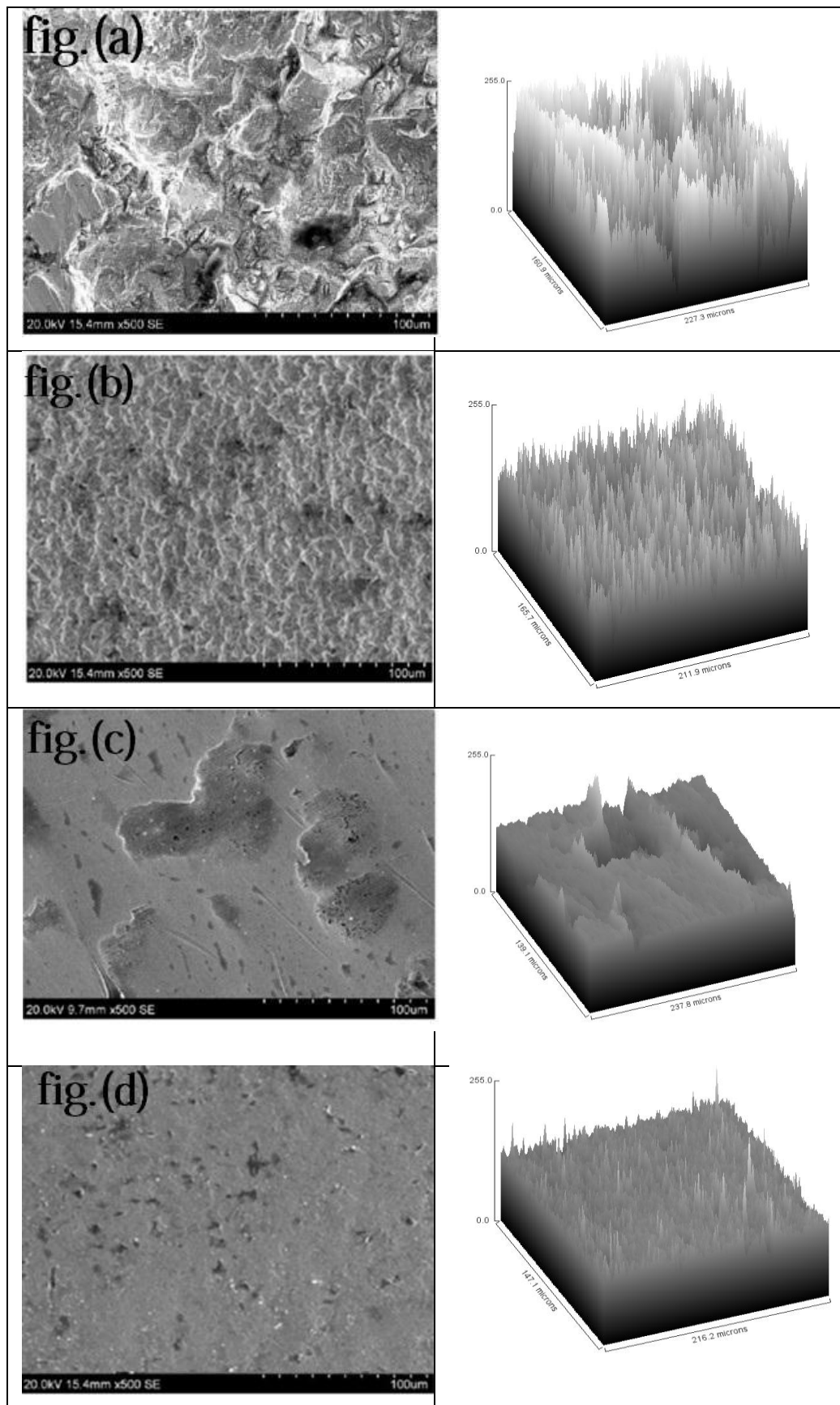
Electrochemical Impedance Spectroscopy (EIS) were obtained for all the Al-2014 alloy samples with inhibitor and without inhibitor in the acidic medium by applying AC signals with amplitude of 10 mV and the measurement was carried out in the frequency range 1MHz–10 mHz at the stable Open Circuit Potential [36].

**3. Results and Discussion****3.1 Microstructure Studies and Elemental Analysis**

The conditions of the metal (Al-2014 alloy) surface were examined using the SEM micrographs after the corrosion tests. The samples of Al-2014 in 1 M HCl solution without and with optimal amoxicillin concentration were given in Figure 2 (a-d). The SEM images depict the differences in the inhibitor-treated surfaces with untreated surfaces. The results can specify that the rate of corrosion decreased drastically with the presence of the inhibitor in the case of the Al-2014 alloy surface. Also, we noticed that the visible pits and cavities in the absence of an inhibitor (figure 2a) were relative to the inhibitor-treated surface (figure 2b). However, the inhibited surface for 50ppm concentration has pits and cracks found, but it is comparatively low with an untreated surface. In the surface analysis, the local erosion of materials leads to a loss in the absence of inhibitors in the acidic medium. When the inhibitor of 50ppm pits and erosion is seen, it is lower than the previous. If the concentration is increased to 100ppm it continues to reduce the pits and erosion, when the 150ppm there is a lower in the pits and erosion due to the creation of a protective inhibitor film because of complete adsorption of inhibitor molecules on the metal surface. It is also visible in figure 2d. Similarly, the surface plots also indicate the same (figure 2 (a-d)). The elemental analysis is carried out by EDAX (figure 3) before the corrosion test and after the measurement; all the elements were noticed and tabulated in Table 1.

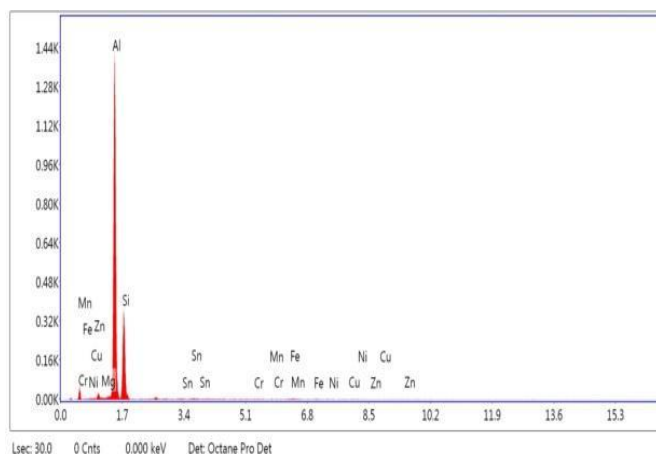
**Table 1: Composition of Al 2014.**

Elements	Mg	Sn	Cr	Mn	Fe	Ni	Si	Cu	Zn	Al
Percentage (Wt. %)	0.22	0.34	0.11	0.16	0.74	0.35	0.8	0.4	0.2	Balance



**Figure 2:** FE-SEM images of the Surface of Al 2014 Alloy (a) without Amoxicillin, and

with Amoxicillin treated surface in 1M HCl medium, (b) 50ppm (c), 100ppm, (D) 150ppm.



**Figure 3:** EDAX of Al-2014 Alloy

### 3.2 Weight loss measurement:

The weight loss measurements are done and estimated the corrosion rate and inhibition efficiency ( $\eta_{WL}\%$ ) of Amoxicillin antibiotic molecules in 1 M HCl on the Al-2014 alloy at 25°C and 35°C. The results are tabulated in Table 2. It is noted that the rate of corrosion depends on the concentration of the Amoxicillin inhibitor. The corrosion rate is high for the untreated surface; it is low for the 100ppm [37].

**Table 2: Corrosion rates of Al-2014 in 1M HCl and Inhibition efficiency for different concentrations of Inhibitor.**

Temperature (°C)	C (ppm)	C <sub>R</sub> in (mpy)	Percentage of efficiency ( $\eta_{WL}\%$ )
25	Blank	3.112	-----
	50	2.613	16%
	100	2.161	30%
	150	1.762	43%
35	Blank	3.963	-----
	50	3.317	15%
	100	2.716	29%
	150	2.096	40%

A similar, trend is also noticed when the temperature is at 35°C. However, the inhibition efficiencies in the 1M HCl medium are more; the corrosion rate is more at 35°C in comparison with 25°C. Because of the phenomenon of acceleration in kinetic reactions, the rate of corrosion generally increases with temperature [38]. Therefore, corrosion efficiency could be reduced. In our study, due to the low-temperature difference (10°C), a small

decrement in the corrosion efficiency was noticed. The appreciable results were reported in the case of mild steel in the acidic medium by Yi Liang et al and they confirmed that the penicillin derivative exhibits the mixed type of inhibitors [39].

### 3.2 Tafel polarization measurement:

Tafel polarization plots as shown in figure 4 and also the details are furnished in table 3. The polarization curves of both anodic and cathodic Tafel change towards lower current density after the addition of Amoxicillin. The  $I_{corr}$  values decrease systematically as the concentration of corrosion inhibitors rises, and the  $\eta_{Tafel}$  values increase, suggesting the formation of a protective film on the Al-2014 surface. The presence of corrosion inhibitors significantly affects the  $E_{corr}$  values for all drug derivatives relative to the uninhibited curve (blank), with the highest  $E_{corr}$  (-662 mV) displacement observed at a concentration of 150ppm for amoxicillin. In General, if the corrosion potential displacement is more than -710 mV concerning the blank corrosion potential. The inhibitor may be considered to be a cathodic or anodic form. Also, the slopes of the cathodic/anodic Tafel lines do not shift periodically by altering inhibitor concentrations, which are also expressed in the relatively stable ' $\beta_c$ ' and ' $\beta_a$ ' [33] values as different from the blank values. Therefore, the drug amoxicillin acts as a corrosion inhibitor, blocking the pathway to reducing corrosion rates at the available metallic active sites. From the plots, we obtained the corrosion efficiency by the following relation (3) [40].

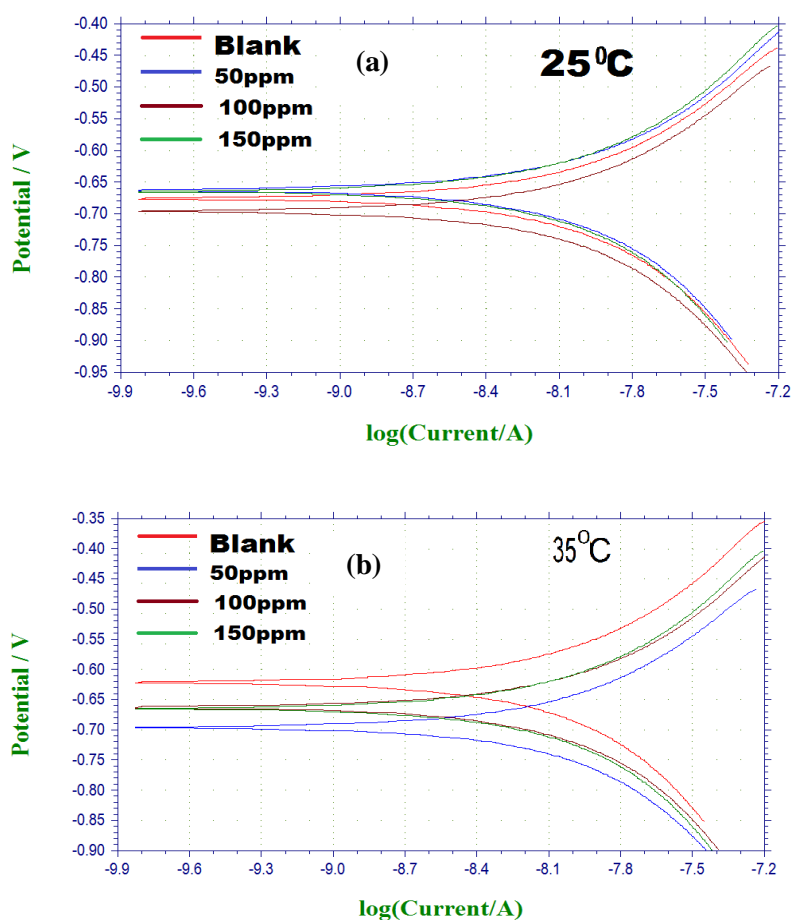
$$\eta_{Tafel} \% = \left(1 - \frac{I_{corr}}{I_{corr}^0}\right) \times 100 \quad (3)$$

Where  $I_{corr}$  and  $I_{corr}^0$  are the corrosion current densities for the Al 2014 alloy immersed in 1 M Solution with and without corrosion inhibitors, respectively.



**Table 3:** Tafel polarization parameters for Al-2014 in 1M HCl solution containing different concentrations of Amoxicillin at different Temperatures.

Temperature ( $^{\circ}\text{C}$ )	Concentration (ppm)	$I_{\text{corr}}$ ( $\mu\text{A cm}^{-2}$ )	$E_{\text{corr}}$ (mV)	$\beta_{\text{c}}$ ( $\text{mV dec}^{-1}$ )	$\beta_{\text{a}}$ ( $\text{mV dec}^{-1}$ )	$\eta$ % Tafel	Surface coverage ( $\theta$ )
25	Blank	1967	-701	107.1	55.3	----	----
	50	1366	-692	111.5	55.9	30%	0.30
	100	755	-670	108.5	54.5	61%	0.61
	150	512	-662	110.2	55.6	73%	0.73
35	Blank	2612	-665	121.1	54.1	----	----
	50	1563	-665	127.8	55.3	40%	0.40
	100	1161	-660	131.9	54.7	55%	0.55
	150	910	-623	124.6	57.3	65%	0.65

**Figure 4:** Tafel polarization curves for Al 2014 alloy in 1.0 M HCl without and with different concentrations of inhibitors at 25°C and 35°C by using Amoxicillin inhibitor.

### a. Adsorption Isotherm

The adsorption isotherm is an important factor to predict the source of the inhibition that directly contributed to the adsorption molecule into the metal surface [41, 42]. The inhibition of molecules that took place due to the adsorption into the metal surface, utilizing physisorption or chemisorption by the electrostatic attraction of charge between metal and molecule or maybe the coordination of electrons and vice versa. Based on the adsorption isotherm factor ( $K_{ads}$ ) we may predict the type of adsorption that leads to the inhibition of molecules. The adsorption isotherm is determined through the following relation (4) [43].

$$\frac{c}{\theta} = \frac{1}{K_{ads}} + C \quad (4)$$

Where: 'C' is the concentration of inhibitor in mole; ' $\theta$ ' is the surface coverage of inhibitor. Similarly, Gibbs free energy ( $\Delta G^0$ ) of the molecule is obtained from the expression (5) [44].

$$\Delta G^0 = -RT \times \ln(55.5 \times K_{ads}) \quad (5)$$

Where; 'R' is the universal gas constant, 'T' is the temperature of the solution medium.

The obtained results are given in table 4. The  $\Delta G^0$  value is negative; it indicates spontaneous inhibition of a molecule into the metal surface by utilizing its molecular energy instead of external energy, and it is found to be in the range of  $-31 \sim -34$  kJ/mole. The obtained value implies that the value is allowed for physisorption ( $<-40$  kJ/mol). Hence, the present case is regarded as the physisorption of the molecule due to the transfer of charge from an organic molecule to the metal surface [45].

**Table 4: Adsorption isotherm factor ( $K_{ads}$ ), And Gibbs free energy ( $\Delta G^0$ ).**

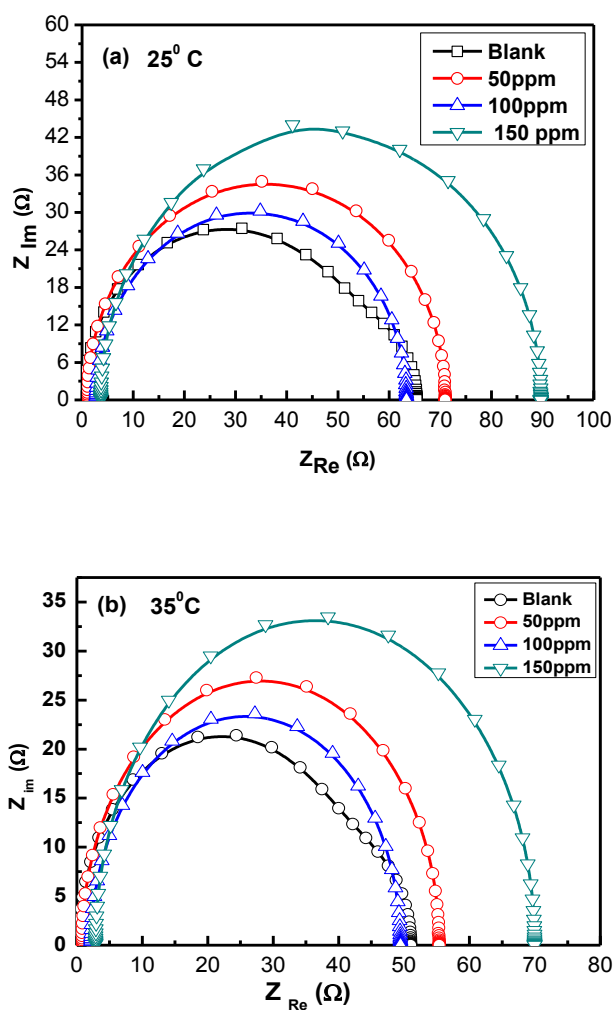
Temperature (°C)	C (ppm)	C (m-mole)	$\theta$	$K_{ads}$ (kJ/mol.)	$\Delta G^0$ (kJ/mol.)
	blank	----	----	----	----
25	50	0.13684	0.30	3283.20	-31.959
	100	0.27370	0.61	5714.20	-33.422
	150	0.41050	0.73	6592.30	-33.799
	Blank	----	----	----	----
35	50	0.13684	0.40	4870.90	-34.108
	100	0.27370	0.55	4465.55	-33.871
	150	0.41050	0.65	4524.09	-33.906

### b. Electrochemical Impedance Studies

Through the cautious assessment of electromagnetic impedance spectroscopy, the obtained values are plotted and shown in figure 5(a & b). The diameter of a semicircle of impedance curve increases with the concentration of inhibitor, it is found to be maximum at 150ppm at a temperature of 25°C as well as 35°C. It is the significance of the inhibition efficiency of Amoxicillin more at 150ppm. If the impedance of the sample is more, it will tend to more actively resist the rate of corrosion by the inhibitor by forming the protective layer that will act as a barrier for charge and mass transfer [46]. Similarly, the diameter of the semicircles is more than that of the sample that has the same concentrations at 25°C. It also infers if the temperature is increased to 35°C the Amoxicillin molecules partially lose the activity to resist corrosion as a result lower the impedance. The examination of the corrosion-resistant layer is carried out by fitting the EIS curves using the Z-Simp software tool, the equivalent circuit is drawn given in figure 6(a & b) and the obtained results are examined and found with the best of 98% accuracy then the details are given in table 5. The critical analysis of data obtained gives that solution resistance ( $R_s$ ) is increased with the concentrations [47]. And also the resistive layer develops the double layer capacitance due to the HOMO and LUMO transition. The oxidative layer is formed in Al-2014 (untreated surface) with all the elements of the alloy, but it accounts for maximum to Al at higher concentrations, the unstable capacitance occurs due to the hooping and oscillating effect of ions by the frequency it is one form of non-ideal behavior of the inhibition barrier, it obeys the relation (6) [48].

$$Z_{CPE} = \frac{1}{Y_0 (j\omega)^n} \quad (6)$$

This result, in developing the inductive effect, for 150ppm, the inductive effect is low and forms the virtually stable resistive layer by the inhibitor. Hence, the corrosion resistance property of Al-2014 increased [49, 50]. The polarization resistance varies due to the unstable secondary layer on the surface of the Al-2014, the metal surface may contain various elements it has different adsorption values hence the inhibition of Amoxicillin is not uniform at low concentrations leads to producing an inductive effect also the presence of dipolar H<sub>2</sub>O in Amoxicillin acts as an oscillating body at high frequency. The inductive effect is significant at 35°C due to the thermal agitation of atoms and molecules in the electrolyte [51].

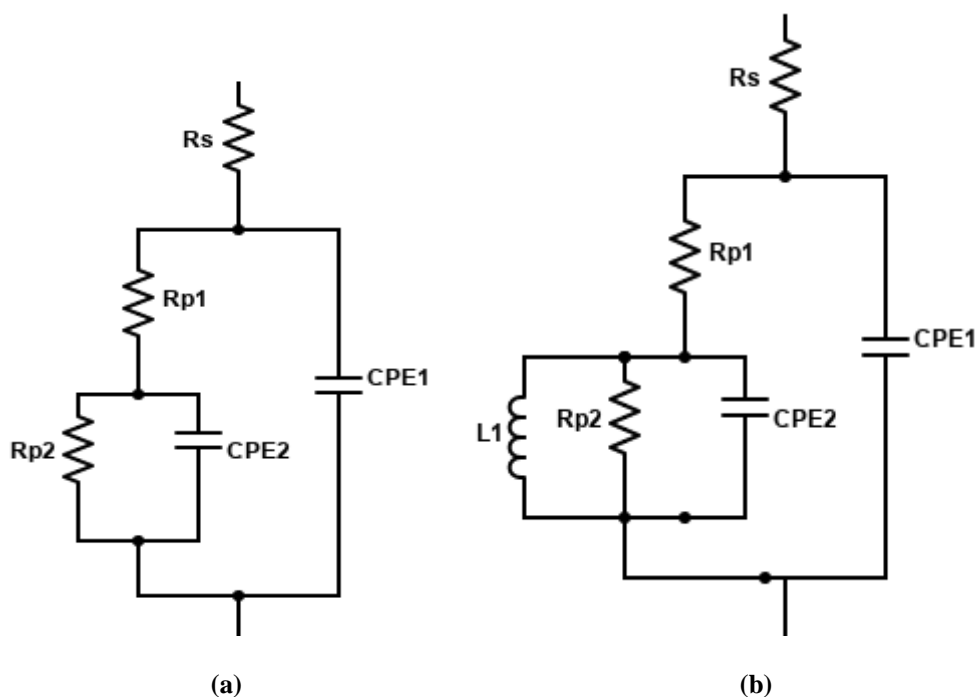


**Figure 5:** Electrochemical impedance spectroscopy for Al-2014 alloy in 1.0 M HCl medium for different concentrations of Amoxicillin inhibitor at 25<sup>0</sup> C and 35<sup>0</sup>C.

**Table 5:** Details of circuit model in EIS, Solution resistance (Rs), Constant phase element (CPE), Polarization resistance (Rp<sub>1</sub> + Rp<sub>2</sub>), Inductance (L), and equivalent Circuit elements

T (°C)	Rs (Ω)	CPE-1 (μF)	Rp <sub>1</sub> (Ω)	CPE-2 (μF)	Rp <sub>2</sub> (Ω)	L (H)	Chi-Squared (x10 <sup>-6</sup> )	Equivalent Circuit
25	0.43	2.5900	63.17	4337	57.80	-----	1.028	R(C(R(CR)))
	0.86	12.820	62.39	4530	65.70	339.4 x10 <sup>-6</sup>	1.053	R(C(R(CRL)))
	1.41	2.0520	45.23	1.511	27.83	1077 x10 <sup>-6</sup>	1.031	R(C(R(CRL)))
	1.83	0.7053	36.46	6624	35.94	3.823 x10 <sup>-8</sup>	2.405	R(C(R(CRL)))
35	0.35	0.1346	42.89	5.603	780.6	-----	1.009	R(C(R(CR)))
	0.80	22.400	54.60	6.33x10 <sup>-3</sup>	1.362x10 <sup>4</sup>	4.010 x10 <sup>-6</sup>	5.125	R(C(R(CRL)))
	2.17	2.3800	47.28	3.27x10 <sup>-2</sup>	1.329x10 <sup>4</sup>	7.822 x10 <sup>-6</sup>	0.991	R(C(R(CRL)))

2.87	1.2580	67.10	13.19	$1.379 \times 10^4$	$1.153 \times 10^{-10}$	1.852	R(C(R(CRL)))
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**Figure 6:** Equivalent circuit model a) without inhibitor-treated surface, b) with the inhibitor-treated surface.

#### 4. Conclusions

The microstructure analysis of Al-2014 alloy with a concentration of 150 ppm implies fewer tendencies to corrosion. It also confirmed from the weight loss method the Percentage of inhibition efficiency  $\eta_{WL}$  is about 43% (25°C) and 40% (35°C). The Tafel polarization was adopted. The following findings are summarized from the obtained results: mix-type inhibitors for Al-2014 corrosion in 1.0 M HCl and the efficiency of inhibition increases as inhibitor concentrations increase. The efficiency of inhibition is as follows: amoxicillin is more at 150ppm 73% (25°C) & 65 % (35°C).also, the adsorption isotherm infers the high rate of inhibition at 150 ppm due to the value of ' $K_{ads}$ '(6592.3kJ/mol.(25°C), 4524.09kJ/mol.(35°C)) and  $\Delta G^0(25^\circ\text{C}, 35^\circ\text{C}) \approx -33.8, -33.9\text{kJ/mol}$ . The same identification is obtained in the Nyquist plots along with the inductive effect are possible for the higher concentrations. The hydrated form of Amoxicillin contains the polar H<sub>2</sub>O molecule. It is concluded that Amoxicillin acts as a better inhibitor on Al-2014 alloy. As the inhibitor has good efficacy and the rate of corrosion also decreases, amoxicillin is a stronger inhibitor than the others in the same derivative of drugs. This inhibitor can be used to avoid corrosion in the oil pipe industry.

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## Declarations

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## Conflict of interest

The authors declared that there was no conflict of interest.

## Data Availability Statement

All data generated or analysed during this study are included in this published article (and its supplementary information files).

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