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Gentamicin, Kanamycin and Amikacin Drugs as Non -Toxic Inhibitors for Corrosion of Aluminum in 1.0M Hydrochloric Acid

M. Abdallah[1,2,*], *B.A.AL Jahdaly*[1]

[1]Chemistry Department, Faculty of Applied Sciences, Umm Al-Qura University, Makkah, Saudi Arabia.
[2]Chemistry Department, Faculty of Science, Benha University, Benha, Egypt
[*]E-mail: metwally555@yahoo.com

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The inhibition effect of three molecules of antibiotic drugs, namely, gentamicin, kanamycin, amikacin on aluminum corrosion in 1.0 M HCl solution has been investigated using weight loss, hydrogen evolution reaction, galvanostatic polarization and electrochemical impedance spectroscopy techniques. The inhibition efficiency increases with increasing the concentration of inhibitor and decreasing temperature. Polarization measurements showed that the studied antibiotic compounds acting as mixed type inhibitors. The adsorption follows the Langmuir isotherm model. Some activated thermodynamic parameters are calculated and explained. One capacitive loop was observed for all spectra in EIS indicating that the corrosion of aluminum corrosion is under charge transfer control.

Keywords: Antibiotic drugs, Aluminum, Corrosion inhibitors, Adsorption, EIS.

1. INTRODUCTION

Aluminum (Al) and its alloys represented as an important class of materials due their high technological value and their wide range of industrial applications, particularly in aerospace, motor vehicle, home industries, pipes, chemical batteries and other studies [1,2]. Acidic solutions are mainly used for chemical and pickling and electrochemical etching of aluminium. It is greatly important to use corrosion inhibitors to decrease the corrosion rate of aluminum in HCl solutions. In most cases, the corrosion inhibitors are mainly organic compounds containing hetero atoms, aromatic rings or multiple bonds [3-18]. It has been known that the first step in the mechanism of inhibition of Al in aggressive HCl solution is generally the adsorption of the organic compounds onto the metal surface. The strength of adsorption process mainly depends on the chemical structure of the adsorbed compound and its

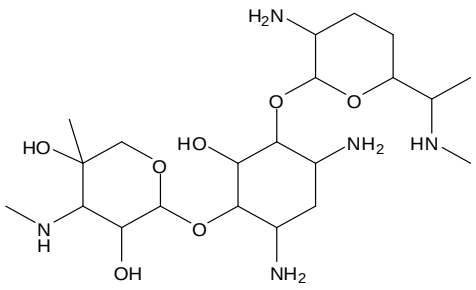
properties such as functional groups, aromaticity and the electron density of the donor atoms-orbital's characters of donating electrons [19]. Thus, development of novel non-toxic corrosion inhibitors have a greet considerable due to their environmental importance [20,21]. In recent years, more attention has been paid from researchers to develop some kinds of drug compounds to be used as corrosion inhibitors for different metals [22-26]. In the previous studies antihypertensive and antibacterial drug compounds were employed as corrosion inhibitors for aluminium in acid media [27,28].

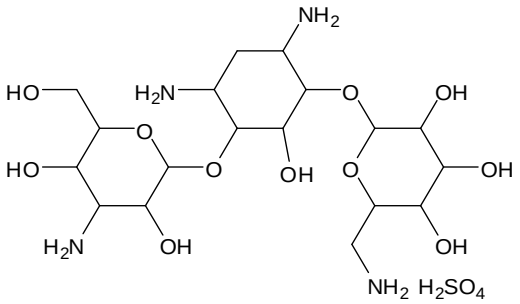
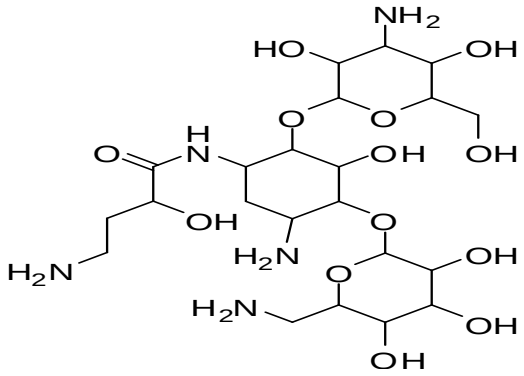
Our work aims to study the effect of three molecules of antibiotic drugs, namely, gentamicin, kanamycin, amikacin toward the aluminum corrosion in 1.0 M HCl solution using weight loss, hydrogen evolution, galvanostatic polarization and EIS methods. The percentage inhibition efficiency was calculated using the above techniques. The effect of rising temperature on the dissolution of Al in 1.0 M HCl solution containing 1000 ppm of antibiotic drugs was studied and some thermodynamic parameters of activation were calculated and discussed.

2. EXPERIMENTAL METHODS

Aluminum with high purity 99. 99% provided by "Aluminum Company of Egypt, Nagh Ammady" was used as working electrode. For weight loss measurements and the hydrogen evolution reaction, the test Al coupons having dimension 2.0x 1.0 x 0.2 cm^3 were abraded with a series of emery papers (grades 320, 600, 800 and 1200) and then washed with bi distilled water and acetone. Finally, dried between two filter papers. The procedure methods of weight loss and hydrogen evolution reaction measurements were carried out as described elsewhere [15,29].

Table 1. The chemical structure of antibiotic drugs

Compounds	Structure
Compound I Gentamicin	 Molecular Formula: $C_{21}H_{43}N_5O_7$ Molecular Weight: 477.59542 g/mol 2-((4,6-diamino-3-((3-amino-6-(1-(methylamino)ethyl)tetrahydro-2H-pyran-2-yl)oxy)-2-hydroxycyclohexyl)oxy)-5-methyl-4-(methylamino)tetrahydro-2H-pyran-3,5-diol

CompoundII Kanamycin	 Molecular Formula: $C_{18}H_{36}N_4O_{11}$ Molecular Weight: 484.49864 g/mol 2-(aminomethyl)-6-((4,6-diamino-3-((4-amino-3,5-dihydroxy-6-(hydroxymethyl)tetrahydro-2H-pyran-2-yl)oxy)-2-hydroxycyclohexyl)oxy)tetrahydro-2H-pyran-3,4,5-triol hydrosulphuric
Compound III Amikacin	 Molecular Formula: $C_{22}H_{43}N_5O_{13}$ Molecular Weight: 585.60252 g/mol 4-amino-N-(5-amino-2-((4-amino-3,5-dihydroxy-6-(hydroxymethyl)tetrahydro-2H-pyran-2-yl)oxy)-4-((6-(aminomethyl)-3,4,5-trihydroxytetrahydro-2H-pyran-2-yl)oxy)-3-hydroxycyclohexyl)-2-hydroxybutanamide

Electrochemical measurements were carried out using Aluminium specimen in the form of a rod of 0.48 cm^2 exposed surface area as a working electrode. The polishing, degreasing and washing of the Al electrode as explained in the above techniques. A PS remote potentiostat with PS6 software to calculate the corrosion parameters was used for the galvanostatic polarization measurements. *EIS* measurements were performed at a frequency range from 10 kHz to 100 mHz and signal amplitude perturbation of 5 mV by using a computer- controlled potentiostate (Auto Lab 30, Metrohm). All the above experiments were performed at 30 ± 1^oC by using ultra circulating thermostat.

The chemical structure of antibacterial drugs is shown in Table 1

3. RESULTS AND DISCUSSION

3.1. Weight loss measurements

Figure 1 represents the weight loss- time relationship for the Al corrosion in 1.0 M HCl solution in the presence and absence of various concentrations of compound (III) at 30±1°C, as a representative example of the studied compounds. Similar curves were obtained for compounds I and II (not shown). The weight loss of Al is decreased. This means that antibiotic molecules slow down the rate of corrosion of Al in 1.0 M HCl solution or in other words, these molecules act as inhibitors.

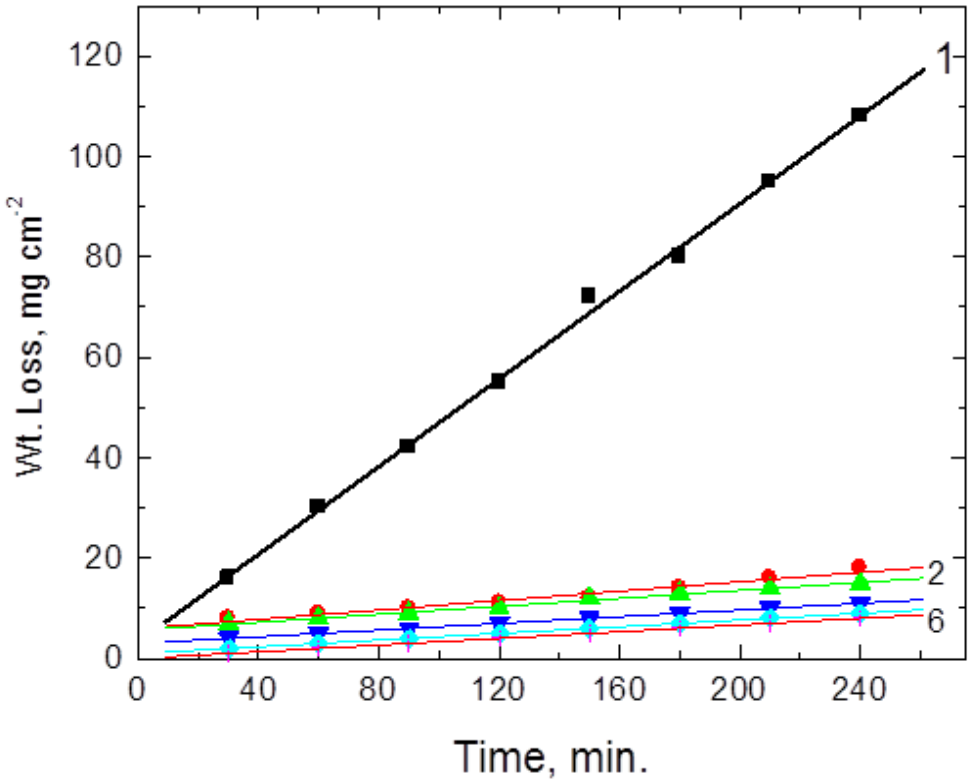


Figure 1. Weight loss-time curves for aluminum dissolved in 1.0 M HCl in the absence and presence of various concentrations of compound III at 30°C. 1- 0.00ppm compound III, 2- 200ppm, 3-400ppm, 4-600ppm, 5-800ppm, 6-1000ppm

As shown by Fig. 1, there is a linear variation of weight loss with time in the absence and presence of inhibitors indicating the absence of insoluble film formation on aluminum surface during corrosion. The investigated compounds are adsorbed first onto the Aluminum surface and thereafter suppress corrosion action either by blocking the reactive sites or by modifying both the cathodic and anodic partial process mechanisms.

The corrosion rate was calculated according to the following relation [30],

$R_{corr} = \Delta W/St$ (1)

where, ΔW(mg) is the weight loss, S is the surface area and t is the immersion time. The surface coverage (θ) and the percentage inhibition efficiency (% IE) were calculated using the following relations:

$$\theta = \left[1 - \frac{R_{add}}{R_{free}}\right] \quad (2)$$

$$\% \text{ I.E} = \left[1 - \frac{R_{add}}{R_{free}}\right] 100 \quad (3)$$

where, R_{free} and R_{add} are the weight loss of Al coupons in the absence and presence of inhibitors, respectively.

The values of R_{corr}., % IE and θ at 30°C are given in Table 2. The values of R_{corr} decrease and the values of % IE increase, indicating the inhibiting effect of the investigated drug molecules and decreases in the following order:

Compound III > Compound II> Compound I

This order will be discussed later.

3.2. Effect of temperature

The effect of increasing temperature on the weight loss of aluminum in 1M HCl solution in the absence and presence of 1000ppm of antibiotic drugs was investigated by weight loss measurements in temperature ranges from (30 to 60°C). Similar curves in Fig. 1 were obtained (not shown). As the temperature rises, the weight loss and the corrosion rate increases and hence the inhibition efficiency, decrease and this is due to the desorption is more easier by increasing the temperature, which indicating that the adsorption of antibiotic drugs on the Al surface occurs through physical adsorption

The activation energy E_a for the corrosion of Al electrode in the absence and presence of 1000 ppm of the antibiotic drugs was calculated using Arrhenius type equation [31,32],

$$\text{Log } R_{corr} = \log A - (E_a/2.303RT) \quad (4)$$

Where, E_a is the apparent activation energy, R is the universal gas constant, A is the Arrhenius pre-exponential factor, T is the absolute temperature

Fig. 2 represents a linear relationship between (log R_{corr}), with (1/T) for Al in 1.0M HCl devoid of and containing 1000 ppm of the antibiotic drugs with slope equal to $-E_a^*/ 2.303R$. The calculated values of E_a obtained from the slope of the straight line are equal to 17.45.18KJ mol^{-1} in 1M HCl and equal 23.78, 24.62 and 25.18 KJ mol^{-1} for compounds I, II and III, respectively.The increase in the activation energy in the presence of antibiotic drugs, indicating the adsorption of the inhibitor molecules on the Al surface making a barrier for mass and charge transfer. The enthalpy and entropy of activation for the corrosion of Al in the absence and presence of drug molecules were calculated using the transition state equation [31,32],

$$\log (R_{corr} / T) = [(\log R/hN) + (\Delta S^*/2.303RT)] - (\Delta H^*/2.303RT) \quad (5)$$

where, h is Planck's constant, N is Avogadro's number, ΔS^* is the entropy of activation and ΔH^* is the enthalpy of activation.

Table 2. Effect of antibiotic drug concentrations on the rate of corrosion ($R_{corr.}$), inhibition efficiency (%I.E) and surface coverage (θ) obtained from weight loss and hydrogen evolution measurements for corrosion of Al in 1.0M HCl solution.

Concentrations	$R_{corr.}$ $mg.cm^{-2}.min.^{-1}$	%IE fromWeight Loss	θ	%IE from H_2 Evolution
1 M HCl + compound I				
00.0ppm compound I	0.443	-	-	-
200ppm compound I	0.096	78.33	0.783	77.45
400 ppm compound I	0.085	80.81	0.808	81.73
600 ppm compound I	0.073	83.52	0.835	83.96
800 ppm compound I	0.061	86.23	0.862	86.89
1000 ppm compound I	0.048	89.16	0.892	90.06
1 M HCl + compound II				
200 ppm compound II	0.087	80.36	0.804	81.10
400 ppm compound II	0.072	83.74	0.837	83.98
600 ppm compound II	0.056	87.53	0.875	88.12
800 ppm compound II	0.045	89.84	0.899	90.41
1000 ppm compound II	0.039	91.19	0.912	91.86
1 M HCl + compound III				
200 ppm compound III	0.067	84.87	0.849	85.04
400 ppm compound III	0.053	87.75	0.877	88.01
600 ppm compound III	.0.044	90.06	0.901	90.56
800 ppm compound III	0.036	91.87	0.919	92.41
1000 ppm compound III	0.028	93.68	0.937	93.12

Fig. 3 represents the relation between of (log R_{corr} /T), with (1/T) for Al in 1.0 M HCl containing 1000 ppm of the studied compounds. Straight lines were obtained with a slope of (-ΔH*/2.303 R) and an intercept [log (R/Nh -ΔS^o /2.303R)]. The values of ΔH* obtained from the slope of the straight line equal to 22.18KJ mol^{-1} in 1M HCl and equal to 25.16, 26.48 and 26.99KJ mol^{-1} in the presence of compounds I, II and III, respectively. These values indicate that, the increase in the enthalpy of activation (ΔH^*) in the presence of the inhibitors implies that the addition of the inhibitors

to the acid solution increases the barrier energy limit of the corrosion reaction to a level depends on the type and concentration of the present inhibitor. The positive values of ΔH* reflect that the adsorption process of the investigated inhibitors on the aluminum surface is an endothermic process

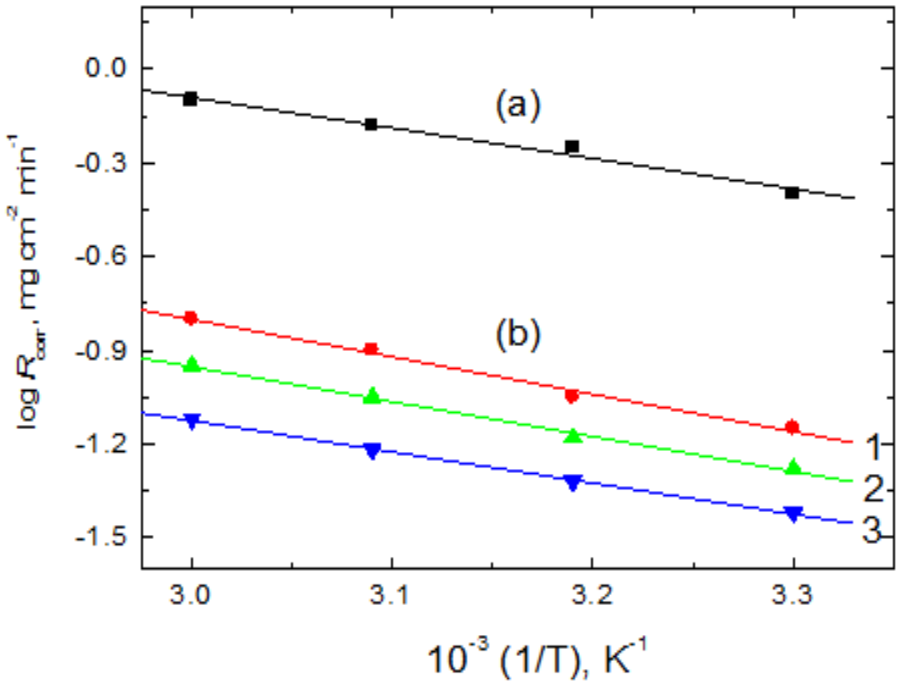


Figure 2. Relation between log R_{corr} and the reciprocal of temperature of Al electrode in a) 1.0 M HCl b) 1.0 M HCl + 1000 ppm of the studied compounds 1)compound I 2) compound II 3)compound III

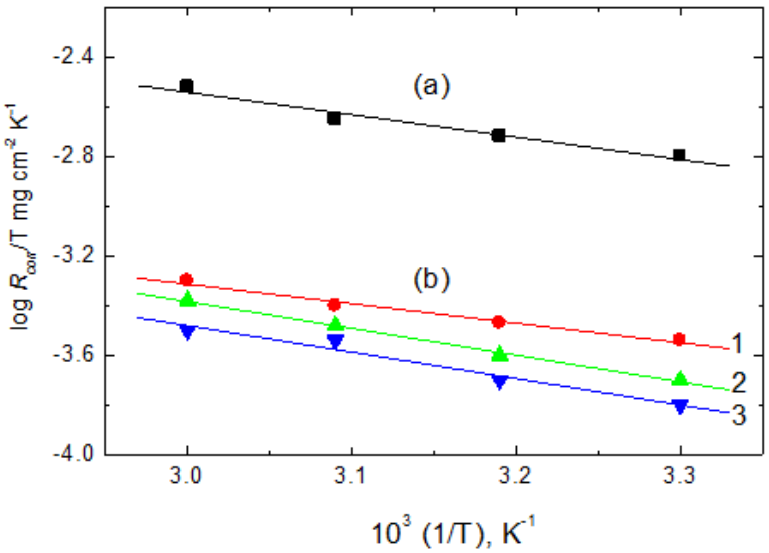


Figure 3. Relation between log R_{corr}/T and the reciprocal of temperature of Al electrode in a) 1.0 M HCl b) 1.0 M HCl + 1000 ppm of the studied compounds 1)compound I 2) compound II 3)compound III

The values of ΔS^* obtained from the intercept of a straight line equal to - 212.34 J mol^{-1} k^{-1} in 1.0MHCl solutions and -243.75 ,254.68 and 258.71 J mol^{-1} k^{-1} in the presence of compounds I, II and III, respectively.

The negative values of ΔS* indicates that the activation complex formation is the rate determining step represents an association rather than dissociation [33].

3.3. Hydrogen evolution measurement

Fig. 4 shows the relation between the volume of hydrogen evolved with time for the dissolution of Al in 1.0 M HCl solution in the absence and presence of various concentrations of compound III as an example of the tested inhibitors.. Similar curves were obtained for other two compounds I and II (not shown). It is clear that from Fig 4. The rate of hydrogen evolution at the beginning of the reaction is low due to the stability of the aluminium oxide (Al_2O_3) film formed at the surface of Al. After certain times the rate of H_2 evolved increases due to start of the dissolution of Al to Al^{+3}. This time is called induction time.

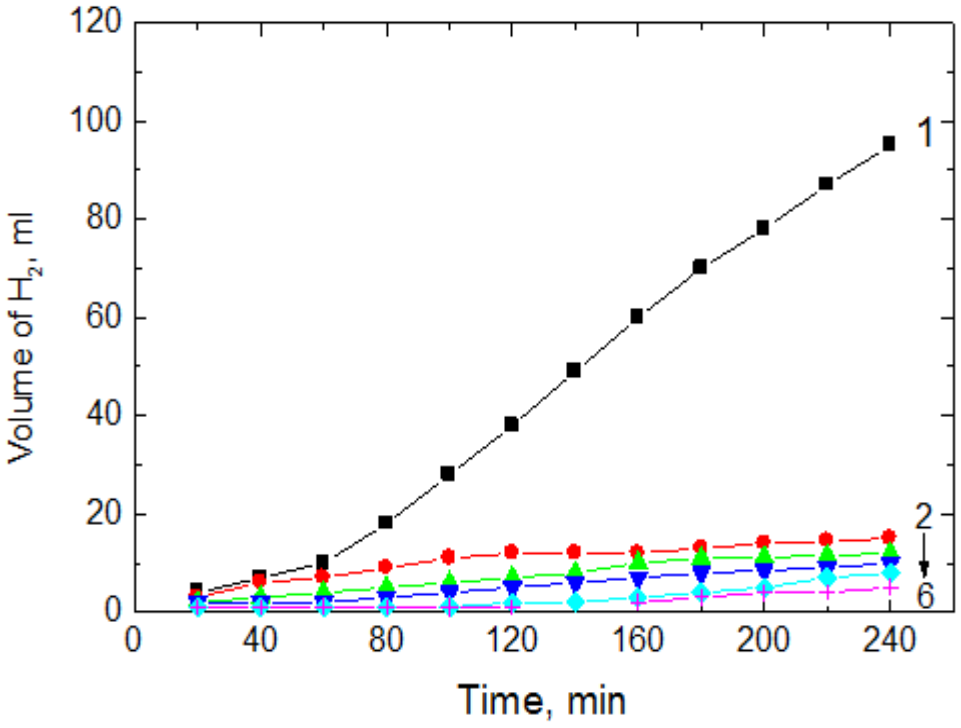


Figure 4. Relation between the volume of hydrogen evolved and time for the corrosion of aluminum in 1 M HCl in devoid of and containing various concentrations of compound III at 30°C. 1-0.00ppm compound III, 2- 200ppm, 3-400ppm, 4-600ppm, 5-800ppm, 6-1000ppm

The corrosion rate was obtained from the slope of the straight portion of curves after induction time. As the concentration of antibiotic drugs increases the rate of H_2 evolved decrease and the

induction time increases, hence the rate of corrosion decreases and the inhibition efficiency increases. This indicates that the inhibiting effect of antibiotic compounds

The percentage inhibition efficiency (% IE) was calculated using the following equations:

$$\% \ IE = [1- R_{free} / R_{add}] 100 \quad (6)$$

where, R_{free} and R_{add} are the corrosion rate of Al coupons in the absence and presence of inhibitors, respectively

The calculated values of (% IE) are given in Table 1. and have the following order: Compound III > Compound II> Compound I

3.4. Galvanostatic polarization

Fig.5. shows the cathodic and anodic polarization curves of Aluminium electrode in 1.0 M HCl solution in the absence and presence of various concentrations of compound III as a representative example of antibiotic molecules. Similar curves were obtained for compounds I, II (not shown).

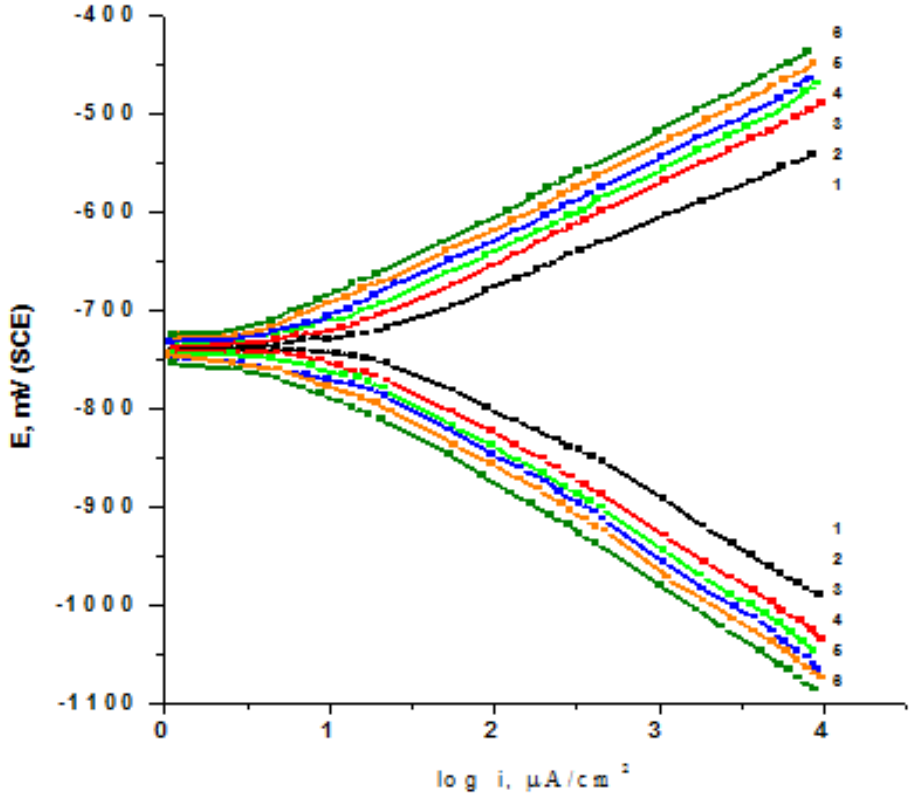


Figure 5. Cathodic and anodic polarization curves of Al electrode in 1.0 M HCl solution in the absence and presence various concentrations of compound III. (1) 0.00ppm compound III, (2) 200ppm, (3) 400ppm, (4) 600ppm, (5) 800ppm, (6) 1000ppm

The corrosion parameters such as, anodic Tafel slope (β_a), cathodic Tafel slope (β_C), corrosion potential ($E_{corr.}$), corrosion current density (I_{corr}), and inhibition efficiency (% IE.) were calculated and given in Table 3.

The percentage inhibition efficiency (%IE) was calculated from corrosion current density values using the equation.

$$\% \text{ IE} = \left[1 - \frac{I_{add}}{I_{free}}\right] 100 \quad (7)$$

where, I_{free} and I_{add} are the corrosion current densities in the absence and presence of inhibitors.As the concentration of antibiotic molecules increases The values of anodic (b_a) and cathodic (b_c)Tafel slopes are nearly constant indicating that these compounds are of mixed type inhibitors. i.e., reduce the anodic and retard the cathodic hydrogen evolution reaction. The values of E_{corr} are shifted to slightly negative potentials and the values of $I_{corr.}$ decreases. Hence the values of %IE increases, indicating the inhibiting effect of the investigated compounds. The calculated values of the percentage inhibition efficiency of antibiotic molecules.decrease in the following order: Compound III > compound II > compound I

Table 3. Corrosion parameters of Al electrode in 1.0 MHCl solution containing various concentrations of of antibiotic drugs.

Inhibitor	Conc., ppm	b_c mV dec^{-1}	b_a mVdec^{-1}	-$E_{corr.}$ mV(SCE)	$I_{corr.}$ µA cm^{-2}	%IE
	0	346	636	748	1258	-
	200	345	637	750	281	77.66
	400	341	638	752	233	81.47
Compound I	600	345	635	753	208	83.46
	800	344	638	756	162	87.12
	1000	344	637	755	112	91.09
	0	346	636	748	1258	-
	200	344	634	746	242	80.76
	400	342	636	748	195	84.49
Compound II	600	348	635	749	152	87.92
	800	347	836	750	118	90.62
	1000	346	633	751	96	92.36
	0	346	636	748	1258	-
	200	345	631	749	199	84.18
	400	344	630	750	145	88.47
Compound III	600	342	632	750	110	91.25
	800	344	633	752	92	92.68
	1000	345	637	753	80	93.64

3.5. Electrochemical impedance spectroscopy (EIS)

Figs. 6 show the Nyquist plots for aluminum electrode in 1.0 M HCl solution in the absence and presence of various concentrations of compound III as a representative example of antibiotic

molecules. Similar curves were obtained for compounds I and II (not shown). The semicircular appearance being observed both in the absence and presence of various concentrations of antibiotic molecules indicating that the corrosion of Al in 1.0 M HCl is mainly controlled by the charge transfer process [34]. The diameter of the capacitive loop increase with increasing the concentration of inhibitors indicating the strengthening of inhibiting film. The impedance spectra of the Nyquist plots (Fig.6) were analyzed by fitting the experimental data to a simple equivalent circuit model which shown in Fig. 7, which includes the solution resistance R_s and the double layer capacitance C_{dl} which is placed in parallel to the charge transfer resistance R_{ct} [35,36].

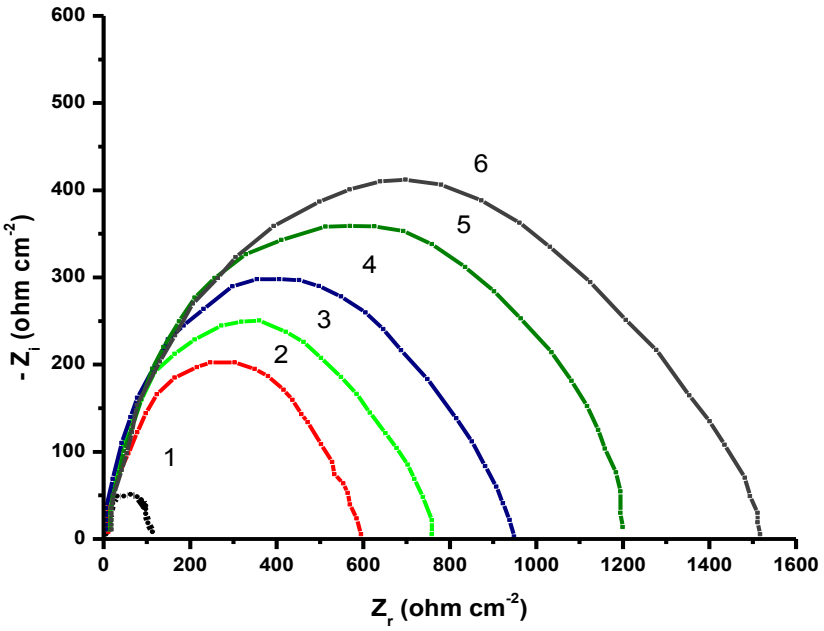


Figure 6. The Nyquist plots for aluminum in 1.0M HCl solution in the absence and presence of various concentrations of compound III at 30^0C 1- 0.00ppm compound III, 2- 200ppm, 3-400ppm, 4-600ppm, 5-800ppm, 6-1000ppm

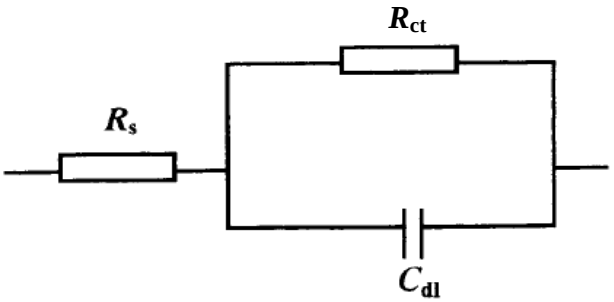


Figure 7. The equivalent circuit model used to fit the experimental results.

The various parameters obtained from the analysis of Nyquist plots are the resistance of charge transfer R_{ct} (diameter of the high frequency loop) and the capacity of double layer C_{dl} which is defined as:

$$C_{dl} = \frac{1}{2\pi f_{max} R_{ct}} \quad (8)$$

Where R_{ct} is the charge transfer resistance in the presence of inhibitor and f_{max} is maximum frequency.

The inhibition efficiency obtained from the impedance measurements is calculated by the following relations:

$$\%IE = \left(1 - \frac{R^{\circ}_{ct}}{R_{ct}}\right) x 100 \quad (9)$$

Where, R^{o}_{ct} and R_{ct} are the charge transfer resistance in the absence and presence of inhibitor, respectively. The associated with the diagrams impedance are given in Table 4.

An inspection of Table 4, as the concentration of antibiotic molecules increases, the values of R_{ct} increases and hence the percentage inhibition efficiency increases due to the formation of adherent film on the Al /solution interface. The values of C_{dl} decrease due to the gradual replacement of water molecule by the adsorption of the antibiotic compounds which form a protective film on the Al surface and led to the decrease in local dielectric constant of the metal solution interface.

The percentage inhibition efficiency obtained from EIS measurements decreases as follows: compound (III) > compound (II) > compound (I)

Table 4. in 1.0 M HCl solution in the absence and presence of various concentrations of antibiotic molecules at 25 ºC. Electrochemical impedance parameters obtained from the Al electrode

Inhibitor	**Conc. (ppm)**	R_s **(Ω cm²)**	R_{ct} **(Ω cm²)**	C_{dl} **(µF cm^{-2})**	**%I.E**
Free	1.0M HCl	8.60	98	25.30	-
	200	9.86	418	23.75	76.55
	400	10.03	498	21.87	80.32
Compound I	600	11.42	715	19.64	86.29
	800	12.86	755	17.84	87.02
	1000	13.98	998	15.66	90.18
	200	10.68	506	23.06	80.63
	400	12.64	576	18.82	82.98
Compound II	600	14.06	695	16.88	85.89
	800	15.68	982	16.22	90.00
	1000	17.86	1085	14.47	90.96
	200	11.55	598	22.50	83.61
	400	13.80	753	19.33	87.03
Compound III	600	15.44	951	18.47	89.69
	800	16.95	1200	15.08	91.83
	1000	18.40	1531	11.50	93.59

The order of inhibition efficiency obtained from the EIS technique were in full accord with those obtained from the weight loss and hydrogen evolution reaction and galvanostatic technique. This proves the validity of these tools in the measurements of the investigated inhibitors.

3.6 Inhibition mechanism and adsorption isotherm

From the results of both chemical and electrochemical measurements obtained above for the corrosion of Al in 1.0 M HCl solution by using some selected antibiotic compounds it was found:

1- The corrosion rate decreases with increasing the concentration of the inhibitor.

2- Weight loss is varied linearly with time.

3- The percentage inhibition efficiency (% IE) decreases with increasing temperature.

4- The rate of hydrogen evolution and the corrosion current density (I_{corr}) decreases with increasing the inhibitor concentration.

5- The values of charge resistance transfer decrease while the double layer capacity increases with increasing the inhibitor concentration.

These results indicate that the inhibition of aluminum corrosion in HCl in the presence of the investigated drug compounds is due to their adsorption at the electrode-solution interface depending on their adsorption active centers, charge density and molecular size [37]. It is generally believed that the adsorption of the antibiotic drugs at the Al / solution interface is the first step in the mechanism of inhibitory action in HCl solution. The adsorption of drug molecules at the metal /solution interface can be elucidated by substitution adsorption between drug molecules in the aqueous solution and the water molecule on the metal surface.

$$Drug_{(sol)} + xH_2O_{(ads)} \longrightarrow Drug_{(ads)} + xH_2O_{(sol)} \quad (10)$$

Where, $Drug_{(sol)}$ and $Drug_{(ads)}$ are drug molecules in solution and adsorbed on the metal surface, respectively. $H_2O_{(ads)}$ is water molecule on metal surface, $H_2O_{(sol)}$ is water molecule in solution and x is the size ratio represent the number of water molecules replaced by one dug molecule.

The inhibition of the investigated antibiotic molecules can be attributed to the presence of heterocyclic rings containing hetero atoms of oxygen and the amino groups ($-NH_2$) in the chemical structure of the studied compounds which led to the formation of more reaction centers for the adsorption. Hence, the adsorption effect of drug molecules could be explained on the basis of the formation of coordination bonds between the unshared electron pairs of oxygen and nitrogen atoms and the empty p-orbits of aluminum. The order of percentage inhibition efficiency (% IE) decreases as follows: compound (III) > compound (II) > compound (I). This is consistent with the molecular weight and the number of hetero atoms (oxygen and nitrogen) within these compounds.

Attempts were made to fit the surface coverage (θ) to various isotherm including Langmuir, Freundlich, Frumkin, Temkin and Flory-Huggins. By far the best results are obtained with Freundlich adsorption isotherm and can be represented using the following equation [38],

$$\theta = KC^n \quad (11)$$

or alternatively by:

$$\log \theta = \log K + n \log C \quad (12)$$

where, K and C are the equilibrium constant of the adsorption process and additive concentration, respectively.

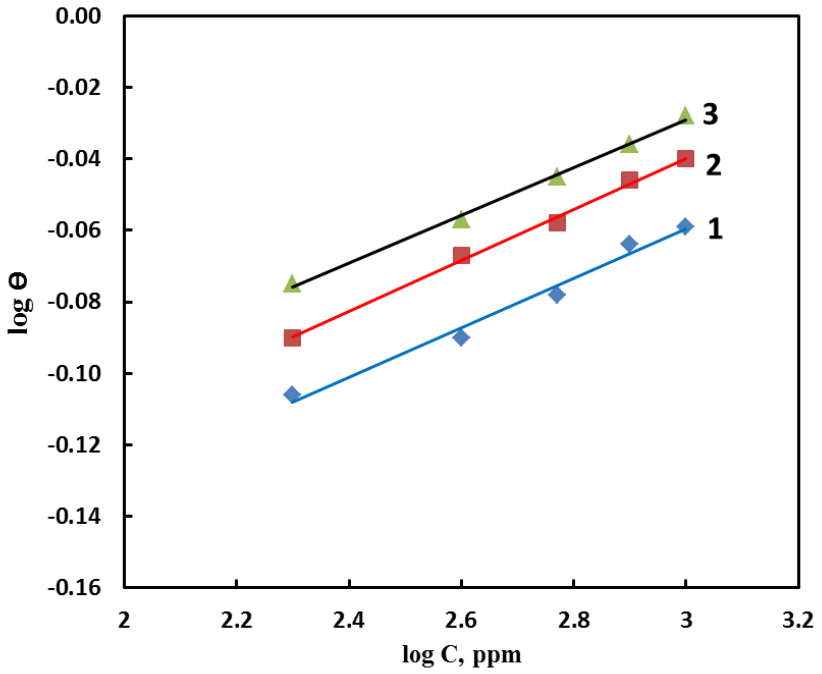


Figure 8. Freundlich adsorption isotherm for Al in 1.0 M HCl solution in the absence and presence of antibiotic drug compounds 1)compound I 2) compound II 3)compound III

Fig 8 represents the plot of log θ versus log C.A straight lines were obtained with intercept of log K.

The equilibrium constant of adsorption, K, is related to the standard free energy of adsorption, ΔG^o_{ads}, according the following equation [23],

$$K=1/55.5 \exp (- \Delta G^o_{ads} /RT) \quad (13)$$

Where, T is the absolute temperature and R is the gas constant (8.314 J. $mol^{-1}.K^{-1}$). The numerical value 55.5 is the concentration of water in $mol.L^{-1}$

The values of K are equal to 0.758, 0.776 and 0.822 for compounds I, II and III, respectively. The high value of K indicates that the strong adsorption of antibiotic drugs on the Al surface. The calculated values of ΔG^o_{ads} for antibiotic drug compounds on the Al surface are equal to –34.56, –36.48 and –38.62kJ mol^{-1} for compounds I, II, and III, respectively.

The free energy change of adsorption is associated with water adsorption/desorption equilibrium which forms an important part in the overall free changes of adsorption. The negative

values of [31,32], ΔG^{o}_{ads} obtained indicates that the adsorption process of antibiotic drugs on the Al surface is spontaneous one.

4. CONCLUSIONS

1-Antibiotic drugs are good inhibitors for the corrosion of aluminum in 1.0 M HCl solution.

2-The inhibition efficiency increases with increasing the concentration of antibiotic drugs and with decreasing temperature.

3-The inhibition process is due to the horizontal adsorption of antibiotic drugs on the Al surface.

4-The adsorption obeys Langmuir isotherm.

5-Antibiotic drugs act as mixed –type inhibitors.

6-EIS measurements indicate that the corrosion reaction is controlled by charge transfer process

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