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Antibacterial drugs as green corrosion inhibitors for copper in hydrochloric acid solutions: Electrochemical and thermodynamic studies

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ABSTRACT

The effect of ampicillin and amoxicillin on the corrosion of copper in 0.1 M HCl was evaluated using potentiodynamic polarization, electrochemical impedance spectroscopy (EIS) and electrochemical frequency modulation (EFM) techniques. The results of polarization studies indicate that the investigated drugs are mixed type inhibitors. It was observed that inhibition efficiency increased with increasing drug concentrations and decreased with raising temperature. Thermodynamic activation and adsorption parameters were calculated and discussed. Adsorption of these drugs on the copper surface was found to follow Langmuir adsorption isotherm. The experimental results are supported by the theoretical data.

Introduction

Copper has an excellent electrical and thermal conductivities, good corrosion resistance and mechanical workability. It is widely used in heating and cooling systems. Corrosion of copper can lead to many problems, the most being per formation that may result in coolant leakage. Scales and corrosion products have negative influence on heat-transfer, causing a decrease in heating efficiencies of the copper structures [1]. Thus, corrosion of copper and its alloys and their inhibition in aqueous chloride solutions have attracted the attention of a member of investigators [2–6]. The use of organic inhibitors is one of the most practical methods for protection against corrosion of metals and their alloys. Generally, numerous organic compounds containing hetero atoms such as nitrogen [7-9], oxygen [10-13], phosphorus [14] and sulphur [15-18] are used as corrosion inhibitors. Studies of the relation between adsorption and corrosion inhibition are of considerable importance. In aqueous solutions, the inhibitory action of organic inhibitors is due to their physical (electrostatic) adsorption onto the metal surface, depending on the charge of the metal surface, the electronic structure of organic inhibitor and the nature of the medium [20, 21].Because of the fact that most of the chemical compounds that prevent the corrosion of metals and alloys are toxic, and thus pose threat both for human health and environment, their usage is limited. For this reason, several authors reported the use of natural products as corrosion inhibitors [19], also, some authors used drugs as green corrosion inhibitors for various metals and alloys [22-30].

The use of environmental friendly pharmaceutical compounds (ampicillin and amoxicillin) as corrosion inhibitors for copper has not been reported before. So, our aim is to study the inhibiting effect of these drugs on copper corrosion in HCl solution using various electrochemical techniques.

Experimental

The working electrode used in the present work was made of pure cylindrical copper rod (99.9 %), welded with copper wire for electrical connection and mounted into suitable glass tube using epoxy resin so that its cross-sectional area (0.5 cm^2) was in contact with the test solution. The exposed area of the electrode was abraded using different grades of emery papers (800- 1200 grade grit), degreased with alcohol, and then washed by bidistilled water and finally dried. The experiments were performed in HCl solution without and with different concentrations of ampicillin and amoxicillin as inhibitors. All solutions were freshly prepared from analytical grade chemical reagents using bidistilled water and were used without further purification (as received).

Potentio dynamic polarization measurements

Polarization experiments were carried out at different temperatures (25-50 °C) in a conventional three-electrode cell with a platinum foil (1 cm^2) as auxilary electrode and a saturated calomel electrode (SCE) coupled to a fine Luggin capillary as reference electrode. The working electrode was in the form of disc cut from copper sheet embedded in epoxy resin of poly tetrafluoroethylene. Before measurement, the electrode was immersed in solution until a steady state was reached (30 min). All experiments were carried out in freshly prepared solutions and results were always repeated at least three times to check the reproducibility. The inhibition efficiency and surface coverage (θ) were determined using equation (1):

$$\%\eta = \theta \text{ x}100 = [1-(i'_{corr}/i_{corr})] \text{ x } 100 \qquad (1)$$

where i_{corr} and i'_{corr} are the current densities in the absence and presence of inhibitor, respectively.

Electrochemical impedance spectroscopy (EIS) measurements

Electrochemical impedance spectroscopy (EIS) was performed at corrosion potential, E_{corr}, over a frequency range of 0^5 Hz to 0.5 Hz with a signal amplitude perturbation of 5 mV. Data were presented as Nyquist and Bode plots. Experiments were always repeated at least three times. Degree of surface

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coverage (θ) and inhibition efficiency (% η) were calculated using equation (2):

$$\% \eta = \theta \times 100 = [(1/R'_{ct})-(1/R_{ct})]/(1/R'_{ct}) \times 100 \qquad (2)$$

where R'_{ct} and R_{ct} are the charge transfer resistance in the presence and absence of inhibitor, respectively.

Electrochemical frequency modulation technique (EFM)

EFM experiments were performed with applying potential perturbation signal with amplitude 10 mV with two sine waves of 2 and 5 Hz. The choice for the frequencies of 2 and 5 Hz was based on three arguments [31]. The larger peaks were used to calculate the corrosion current density (i_{corr}), the Tafel slopes (b_c & b_a) and the causality factors CF-2 and CF-3 [32, 33]. The inhibition efficiency % η_{EFM} was calculated as follows:

$$\% \eta_{EFM} = [1- (i_{corr} / i^{o}_{corr})] \times 100 \qquad (3)$$

All the electrochemical experiments were carried out using Gamry PCI300/4 Potentiostat / Galvanostat/Zra analyzer, EIS300 electrochemical impedance software, EFM140 electrochemical frequency modulation software, DC105 polarization software and Echem 5.21 for results plotting, graphing, data fitting and calculating.

Quantum chemical calculations

Materials studio V.4.4.0 was used for molecular modeling .The molecular orbital calculation are based on a semiempirical self-consistent field molecular orbital (SCF-MO) method A full optimization of all geometrical variables without any symmetry constraints was performed at the restricted Hartee–Fock (RHF) level using Parameterization Model 3 (PM3) method.

Results and discussion

Potentiodynamic polarization measurement

Potentiodynamic polarization curves of copper electrode in 0.1 M of HCl solution without and with various concentrations of ampicillin and amoxicillin at different temperatures (25–55◦C) were recorded. The polarization profile of copper in 0.1 M HCl at 25°C in the presence of increasing amounts of amoxicillin is shown in Fig. 1. Similar curves were obtained for ampicillin in Fig. 2. Electrochemical parameters such as corrosion current density (i_{corr}), corrosion potential (E_{corr}), anodic (b_a) and cathodic (b_c) Tafel slopes in all cases were calculated from Tafel plots. The calculated values are listed in Table 2. The presence of increasing amounts of amoxicillin led to a decrease in both the cathodic and anodic current density. This behavior was observed for all of the temperatures under study. It is clear from these data that the inhibitors may affect either the anodic or the cathodic reaction, or both [34]. Since the anodic (b_a) and cathodic (b_c) Tafel slopes of ampicillin and amoxicillin were found to change with inhibitor concentration, this indicates that these inhibitors affected both of these reactions [35].

The addition of ampicillin and amoxicillin shifts the E_{corr} values towards the negative. A compound can be classified as an anodic-or cathodic-type inhibitor when the change in E_{corr} value is larger than 85 mV [36]. Since the largest displacement exhibited by ampicillin and amoxicillin was less than 85 mV (Table 2), it may be concluded that these molecules should considered as a mixed-type inhibitor, meaning that the addition of these drugs to 0.1 M HCl solution both reduces the anodic dissolution of copper and also retards the cathodic hydrogen evolution reactions. From the calculated values of (% η) at different temperatures as shown in Table 2, the order of decreasing inhibition efficiency is: ampicillin > amoxicillin.

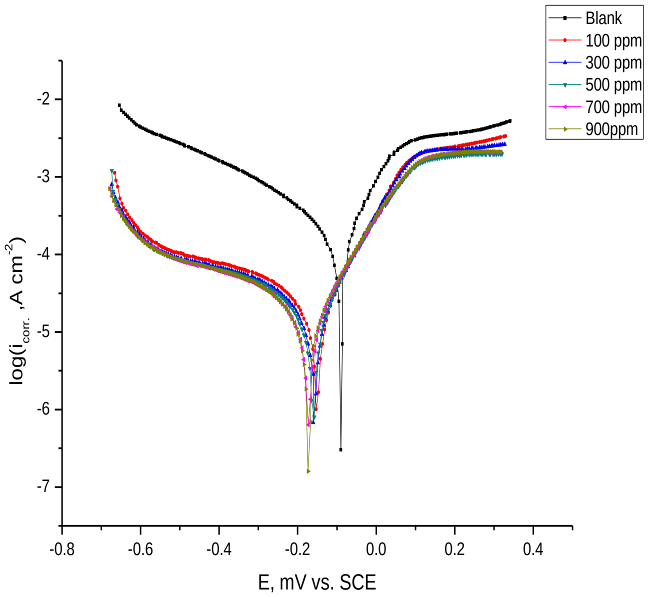


Figure 1. Potentiodynamic polarization curves of copper in 0.1 M HCl at different concentrations of amoxicillin at 25°C

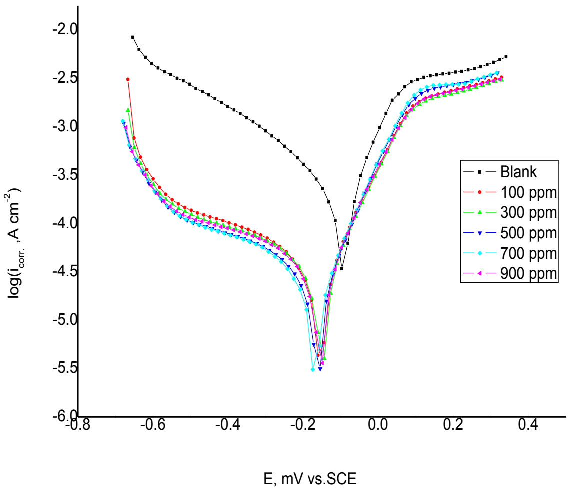
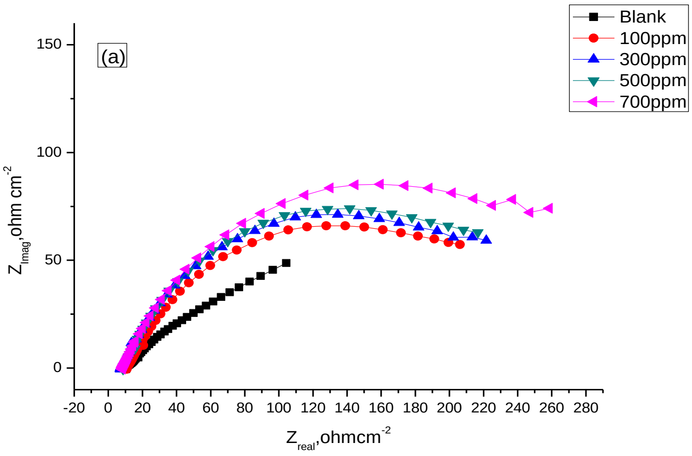


Figure 2. Potentiodynamic polarization curves of copper in 0.1 M HCl at different concentrations of ampicillin at 25°C



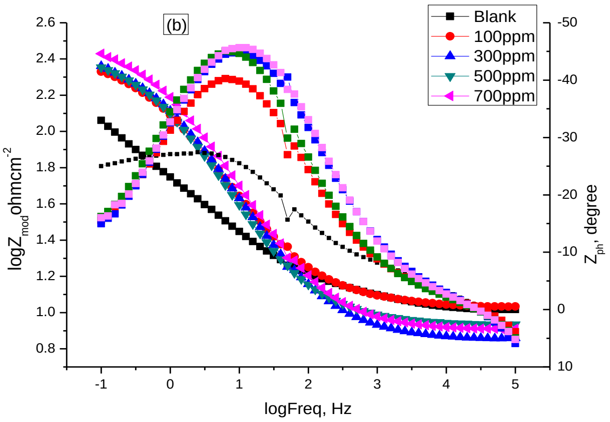


Figure 3. Nyquist (a) and the Bode (b) plots for corrosion of copper in 0.1 M HCl in the absence and presence of different concentrations of amoxicillin at 25°C

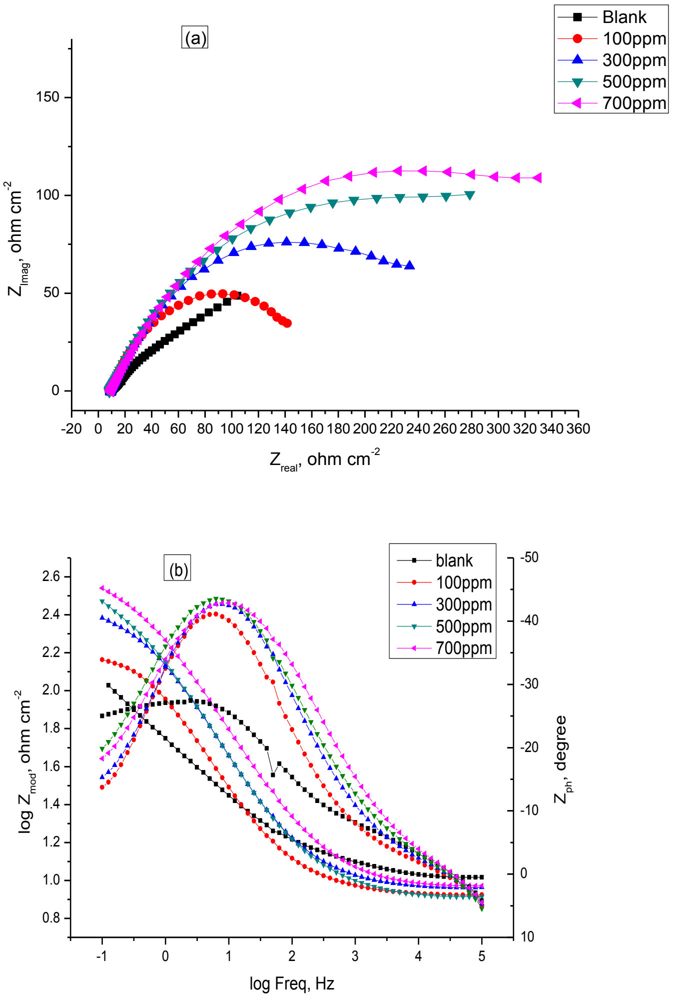


Figure 4.The Nyquist (a) and Bode (b) plots for corrosion of copper in 0.1 M HCl in the absence and presence of different concentrations of ampicillin

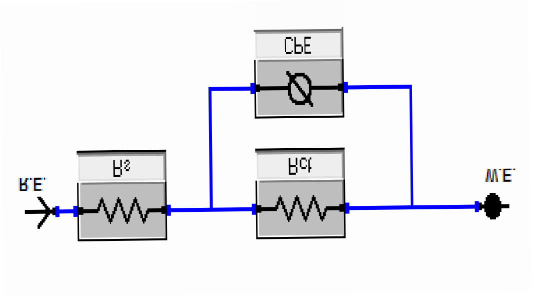


Figure 5. Equivalent circuit proposed to fit the EIS experimental data

Electrochemical impedanc spectroscopy (EIS) measurements

EIS is a well-established and powerful technique in the study of corrosion. Surface properties, electrode kinetics and mechanistic information can be obtained from impedance diagrams. Figure 3 (a& b) shows the Nyquist and Bode plots obtained in the absence and presence of increasing concentrations of amoxicillin. Similar curves were obtained for ampicillin Figure 4 (a&b). The Nyquist plots of ampicillin and amoxicillin are not perfect semicircles, which is attributed to nonhomogeneity of the surface and roughness of the metal [37]. From the plots, it could be seen that impedance response of copper is increased by the addition of ampicillin and amoxicillin [38]. For a corrosion system, the formation of double layer at metal/solution interface can be represented by the electronic equivalent circuit (Figure 5).

The double layer capacitance (C_{dl}) and the charge transfer resistance (R_{ct}) values calculated from Nyquist plots are listed in Table 3. The R_{ct} values increased with the increase in the concentration of inhibitors, which shows protection of copper surface by the inhibitors while the values of C_{dl} decreased with the increase in the concentration of inhibitors, which is due to the increase in the thickness of protective layer at higher concentrations [39, 40]. Inhibition efficiency, calculated from the values of R_{ct} [41] was found to be maxima at a concentration of 700 ppm of both inhibitors. The results of EIS were in good agreement with the results of polarization. It was found that the inhibition efficiency (% η_{EIS}) of these inhibitors follows the following sequence: amoxicillin > ampicillin.

Electrochemical frequency modulation (EFM) measurements

EFM is a nondestructive corrosion measurement technique that can directly give values of the corrosion current without a prior knowledge of Tafel constants. The great strength of the EFM is the causality factors which serve as an internal check on the validity of EFM measurement. The causality factors CF-2 and CF-3 are calculated from the frequency spectrum of the current responses shown in Table 4. Fig 6 shows the frequency spectrum of the current response of copper in HCl solution. From the results of Table 4, the inhibition efficiency (%η_{EFM}) of these drugs follows the same sequence as before: amoxicillin > ampicillin.

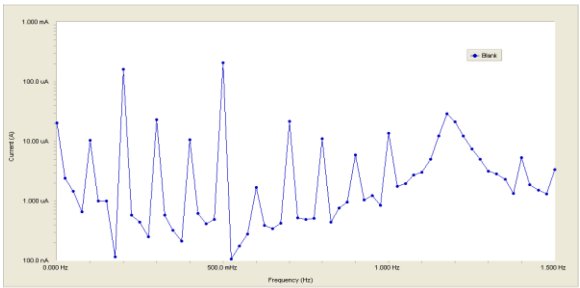


Figure 6. Intermodulation spectra for copper in absence of drugs

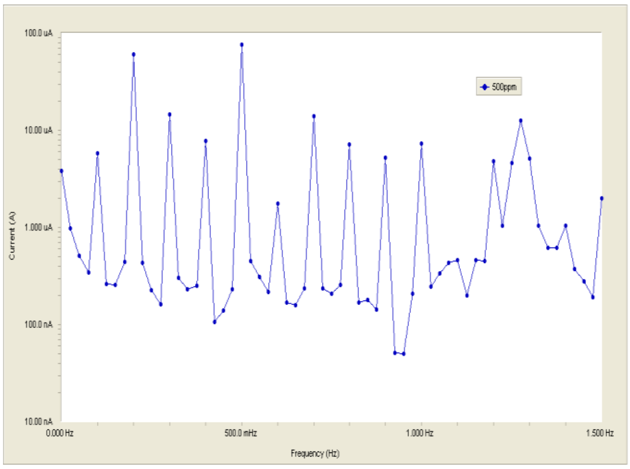


Figure 7: Intermodulation spectra for copper in 500 ppm of ampicillin

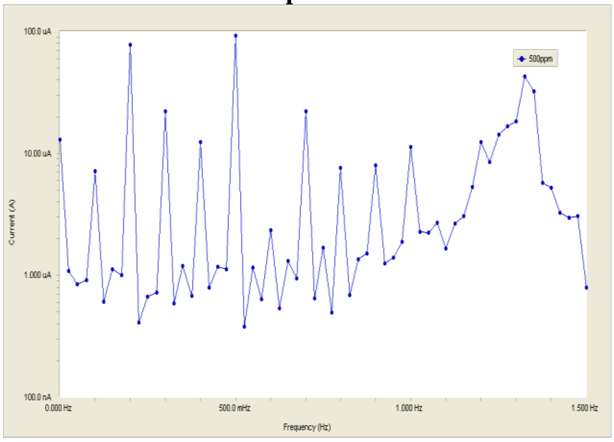


Figure 8. Intermodulation spectra for copper in 500 ppm of amoxicillin

Adsorption isotherms

The investigated compounds inhibit the corrosion by adsorption at the metal surface. Theoretically, the adsorption process has been regarded as a simple substitutional process, in which an organic molecule in the aqueous phase substitutes an (y) number of water molecules adsorbed on the metal surface.

A number of mathematical relationships for the adsorption isotherms have been suggested to fit the experimental data of the present work. The simplest equation is that due to [42] Langmuir which is given by the general relation:

$$C / \Theta = (1/ K_{ads}) + C \qquad (4)$$

where K_{ads} is the equilibrium constant for the adsorption/desorption process, C is the inhibitor concentration in the bulk of the solution in mol L^{-1}.

From the intercepts of the straight lines on the C/Θ axis, (Figs. 9 and 10), one can calculate K_{ads} values that relate the standard free energy of adsorption, ($\Delta G°_{ads}$), as given by Eq. (5) [43]:

$$K_{ads} = 1/55.5 \exp (-\Delta G°_{ads}) /RT \qquad (5)$$

The value of 55.5 is the concentration of water in the bulk solution in mol L^{-1}.Free energy, $\Delta G°_{ads}$, values were calculated and are given in Table 5, the negative values of $\Delta G°_{ads}$ indicate spontaneous adsorption of inhibitors on copper surface [44]. Generally, values of $\Delta G°_{ads}$ up to -20 kJ mol^{-1} are consistent with physisorption, while those around -40 kJ mol^{-1} or higher are associated with chemisorptions as a result of the sharing or transfer of electrons from inhibitor molecules to the metal surface to form co-ordinate bond [45]. The calculated $\Delta G°_{ads}$ values are 23-35 kJ mol^{-1} indicating that the adsorption mechanism of amoxicillin and ampicillin in 0.1 M HCl solution at the studied temperatures is mixed one (physisorption and chemisorption).

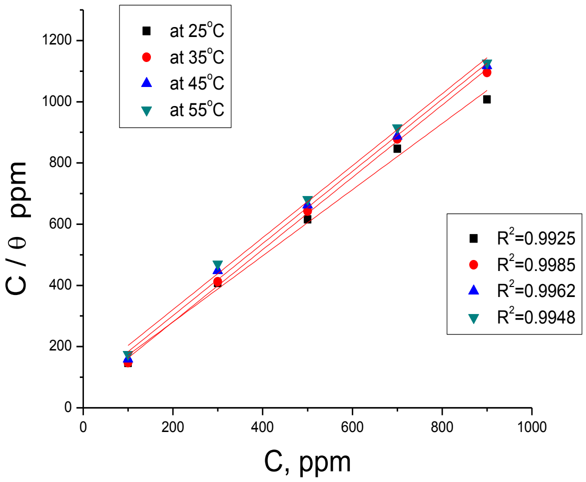


Figure 9. The linear form of Langmuir adsorption isotherm of ampicillin on copper surface in 0.1 M HCl solution

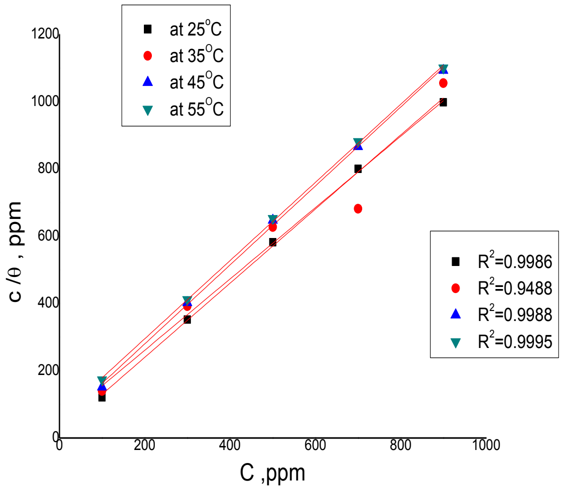


Figure 10. The linear form of Langmuir adsorption isotherm of amoxicillin on copper surface in 0.1 M HCl solution

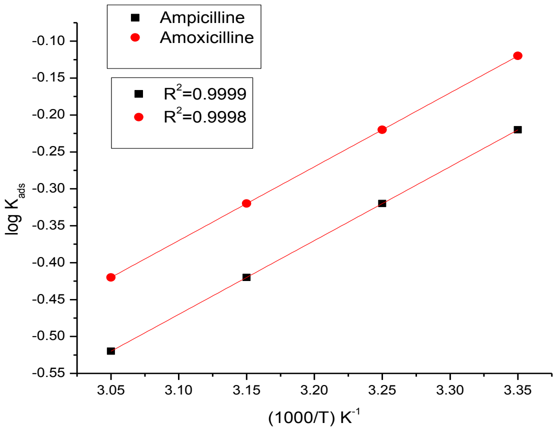


Figure 11. log Kads vs. (1/T) curves for the corrosion of copper in 0.1 M HCl in the absence and presence of different concentrations of amoxicillin and ampicillin at different temperatures

Table 1: Name and the structural formula of the investigated inhibitors

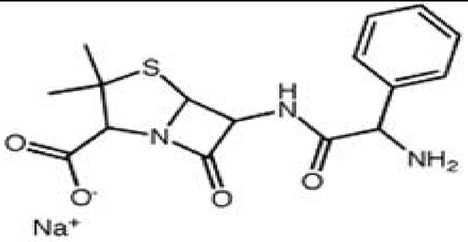
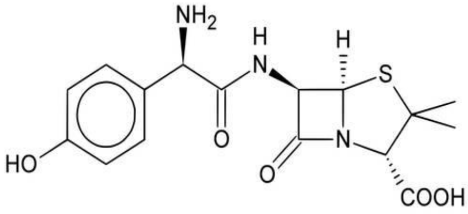
Name	**Structure**	**Mol. Wt.**
Ampicillin Sodium6-(2-amino-2-phenyleacetamido)-3,3-dimethyle-7-oxo-4-thia-1-azobicyclo[3.2.0]heptane-2-carboxylate.	 $C_{16}H_{19}N_3O_4S$	371.39
Amoxicillin 6-(2-amino-2-(4-hydroxyphenyl) acetamido)-3,3-dimethyl-7-oxo-4-thia-1-azabicyclo[3.2.0]heptane-2-carboxylic acid.	 $C_{16}H_{19}N_3O_5S$	419.45

Table 2: Electrochemical parameters obtained from potentiodynamic polarization measurements for copper in 0.1 M of HCl in the absence and presence of different concentrations of ampicillin and amoxicillin at 25°C

	Conc. ppm	**-E_{corr}, mVvsSCE**	**i_{corr} μA cm^{-2}**	**b_c, mV dec^{-1}**	**b_a, mV dec^{-1}**	**R_P Ohm cm^{-2}**	θ	**%η**	**υ mmy^{-1}**
Blank		94	284.10	670	351	352	----	----	3.296
Ampicillin	100	141	89.91	730	371	903	0.684	68.4	1.373
	300	150	74.89	1325	261	1265	0.736	73.6	0.869
	500	160	53.42	951	232	1513	0.812	81.2	0.620
	700	167	49.10	841	229	1593	0.827	82.7	0.570
	900	153	30.36	523	221	2223	0.893	89.3	0.352
Amoxicillin	100	151	48.18	920	231	1661	0.831	83.1	0.559
	300	159	41.93	876	237	1933	0.853	85.3	0.486
	500	160	40.91	897	247	2054	0.859	85.9	0.475
	700	167	35.68	814	246	2210	0.874	87.4	0.414
	900	174	27..93	592	227	2554	0.902	90.2	0.324

Table 3: Electrochemical kinetic parameters obtained from EIS technique for copper in 0.1 M HCl in the absence and presence of different concentrations of ampicillin and amoxicillin

Comp.	**Conc. ppm**	**R_p ohm cm^2**	**C_{dl} μFcm^{-2}**	**Ɵ**	**% η**
Blank	----	172.7	1560	----	----
Ampicillin	100	253.1	1220	0.318	31.8
	300	261.2	1190	0.339	33.9
	500	262.8	1030	0.343	34.3
	700	303.5	835	0.431	43.1
Amoxicillin	100	285.7	1490	0.395	39.6
	300	297.9	1320	0.422	42.2
	500	390.0	1090	0.557	55.7
	700	447.0	963	0.614	61.4

Table 4. Electrochemical kinetic parameters obtained by EFM technique for copper in 0.1 M HCl in the absence and presence of different concentrations of ampicillin and amoxicillin at 25 °C

Comp.	Conc, ppm	i_{corr} μA cm^{-2}	b_a mV dec^{-1}	b_c mV dec^{-1}	CR mpy	CF-2	CF-3
Blank		135.00	40	57	34.66	1.828	3.045
Ampicillin	100	91.71	32	69	23.99	1.977	3.127
	300	55.95	34	74	14.64	1.941	3.167
	500	46.37	34	62	12.13	1.910	3.056
	700	41.82	37	74	10.94	1.921	3.005
Amoxicillin	100	66.72	37	74	17.45	1.900	3.248
	300	53.65	28	50	12.19	2.108	4.547
	500	46.60	31	62	14.04	1.865	3.581
	700	40.48	29	53	10.59	1.759	3.417

Table 5. Thermodynamic parameters for the adsorption of ampicillin and amoxicillin on copper in 0.1 M HCl at different temperatures

Inhibitors	Temp., K	K_{ads}x10^{-4} M^{-1}	$-\Delta G^{\circ}_{ads}$ kJ mol^{-1}	ΔH°_{ads}- kJ mol^{-1}	ΔS°_{ads}- K^{-1} J mol^{-1}
Ampicillin	298	0.60	29.15	21.2	76.0
	308	0.42	26.18		
	318	0.41	25.98		
	328	0.31	23.66		
Amoxicillin	298	1.22	35.05	41.3	137.9
	308	0.52	27.96		
	318	0.48	27.29		
	328	0.38	25.35		

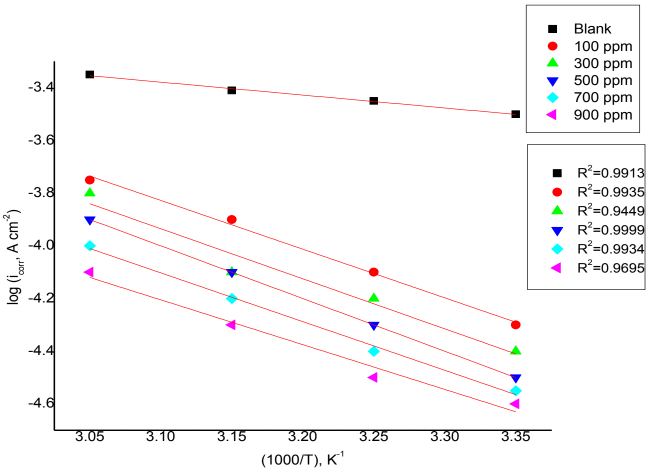


Figure 12. log i_{corr} vs. (1/T) curves for the corrosion of copper in 0.1 M HCl in the absence and presence of different concentrations of ampicillin at different temperatures

Thermodynamic adsorption parameters

Thermodynamic adsorption parameters such as enthalpy of adsorption ΔH°_{ads} and entropy of adsorption ΔS°_{ads} can be deduced from integrated version of the Vant't Hoff equation expressed by [46]:

$$\ln K_{ads} = (-\Delta H^{\circ}_{ads}/RT) + (\Delta S^{\circ}_{ads}/R) + \ln (1/55.5) \qquad (6)$$

Figure 11 shows the plot of ln K_{ads} vs. 1/T which gives straight lines with slopes $(-\Delta H^{\circ}_{ads}/2.303R)$ and intercepts $[(\Delta S^{\circ}_{ads}/2.303R) + \ln (1/55.5)]$. Calculated values of ΔH°_{ads} and ΔS°_{ads} using the Van't Hoff equation are listed in Table 5. The negative sign of ΔH°_{ads} indicates that the adsorption of amoxicillin and ampicillin on copper surface is an exothermic process. The average value of ΔH°_{ads} is about 20-40 kJ mol^{-1} which is larger than the common physical adsorption heat (40 kJ mol^{-1}), but smaller than the common chemical adsorption heat (100 kJ mol^{-1}) [47], probably meaning that both physical and chemical adsorption take place (i.e. comprehensive adsorption).

The negative ΔS°_{ads} values are accompanied with exothermic adsorption process. This is agrees with the expected, when the adsorption is an exothermic process, it must be accompanied by a decrease in the entropy change and vice versa [48].

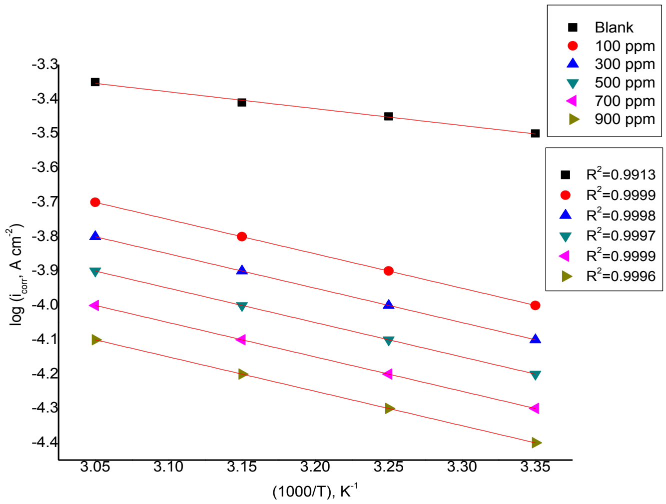


Figure 13. log i_{corr} vs. (1/T) curves for the corrosion of copper in 0.1 M HCl in the absence and presence of different concentrations of amoxicillin at different temperatures

Effect of temperature

Generally the corrosion rate increases with the rise of temperature. It was found that the inhibition efficiency decreases with increasing temperature. This can be attributed to the shift of the adsorption–desorption equilibrium towards desorption. Such behavior suggests that investigated compounds were physically adsorbed on copper surface. The activation energy (E^*_a) of the corrosion process was calculated using Arrhenius equation:

$$i_{corr} = A \exp(-E^*_a/RT) \qquad (7)$$

where A is Arrhenius constant, R is the gas constant and T is the absolute temperature.

Figures12 and 13 show the Arrhenius plot (log i_{corr} vs. 1/T) in the presence and absence of ampicillin and amoxicillin. The values of activation energies E^*_a can be obtained from the slopes of the straight lines and are given in Table 6.

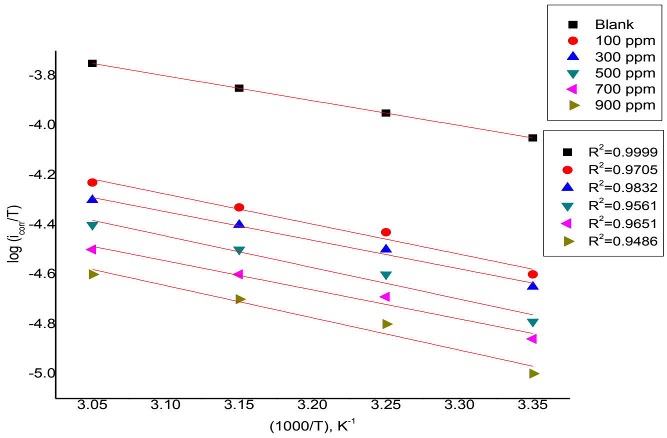


Figure 14. log (i_{corr}/T) vs. (1/T) curves for the corrosion of copper in 0.1 M HCl in the absence and presence of different concentrations of ampicillin at different temperatures

It is noted that the values of activation energy is higher in the presence of inhibitors than in their absence indicating that inhibitors exhibit low % η at elevated temperatures [49] and also, due to the film formation on copper surface. An alternative formulation of the Arrhenius equation is the transition state equation [50]:

$$i_{corr} = RT/Nh \exp(\Delta S^*/R)\exp(-\Delta H^*/RT) \qquad (8)$$

where h is Planck's constant, N is Avogadro's number, ΔS^* is the entropy of activation and ΔH^* is the enthalpy of activation. Fig. 14,15 shows a plot of log (i_{corr}/T) vs. (1/T). Straight lines were obtained with slopes of ($\Delta H^*/2.303R$) and intercepts of (log R/ Nh +$\Delta S^*/2.303R$) from which the values of ΔH^* and ΔS^* were calculated and listed in Table 6. The negative values of ΔH^* reflect that the process of corrosion is an exothermic one. The negative values of ΔS^* implies that the activation complex is the rate determining step that represents an association rather than dissociation step [51]. This means that the activated molecules are in higher order state than that the initial state.

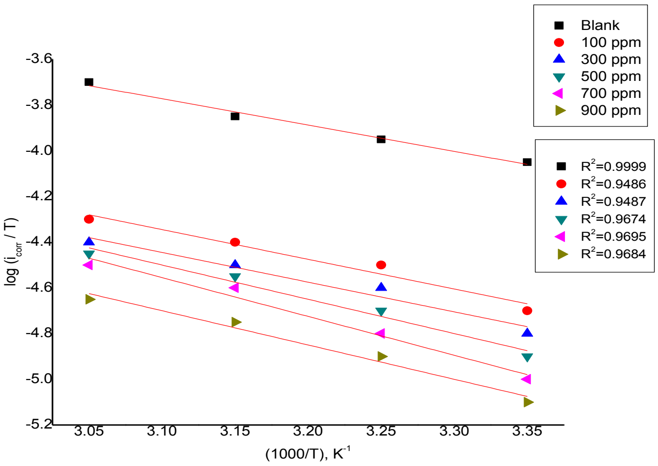


Figure 15. log (i_{corr} /T) vs. (1/T) curves for the corrosion of copper in 0.1 M HCl in the absence and presence of different concentrations of amoxicillin at different temperatures

Quantum chemical calculation

Semi empirical methods serve as efficient computational tools which can yield fast quantitative estimates for a number of properties [52]. MNDO (modified neglect of differential overlap) is a semiempirical methods based on the NDDO (neglect of diatomic differential overlap) approximation.

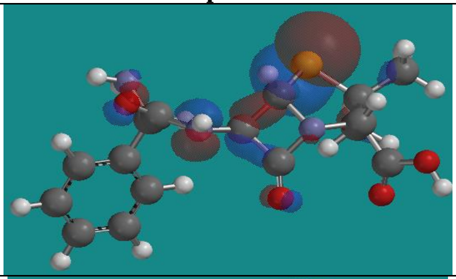
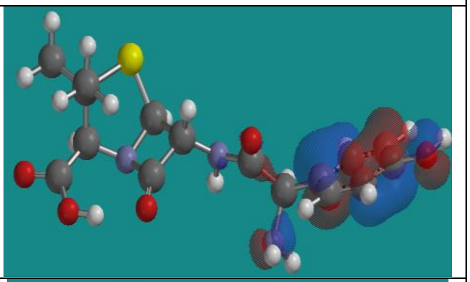
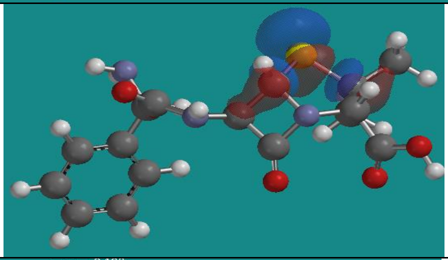
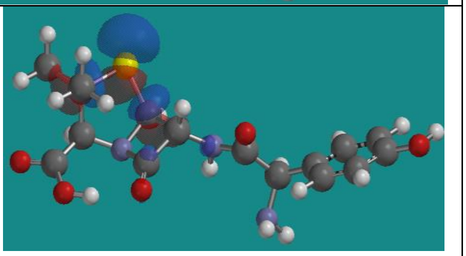
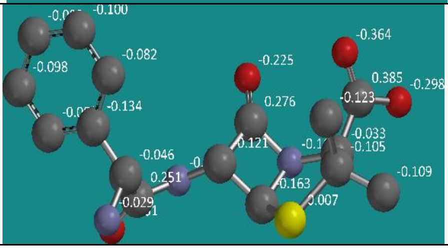
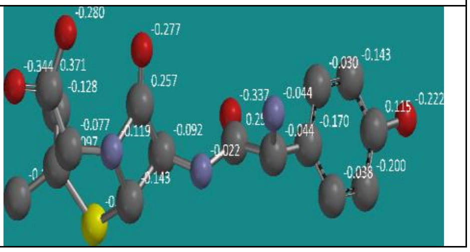
	ampicillin	**Amoxicillin**
HOMO		
LUMO		
Mulliken charge		

Figure 16. Molecular orbital plots and Mulliken charge of ampicillin and amoxicillin using MNDO

Several researchers have shown that the adsorption of an inhibitor on the metal surface can occur on the basis of donor-acceptor interaction between the p-electrons of the heterocyclic compound and the vacant d- orbital of the metal surface atoms [53-55]. According to the frontier molecular orbital theory, the formation of a transition state is due to an interaction between the frontier orbitals (HOMO and LUMO) of reactants [56]. E_{HOMO} is often associated with electron donating ability of the molecule. High E_{HOMO} values indicate that the molecule has a tendency to donate electrons to appropriate acceptor molecules with low energy empty molecular orbital. Increasing values of the E_{HOMO} facilitate adsorption by influencing the transport process through the adsorbed layer [57&58]. The higher the HOMO energy level of the inhibitor, the greater is the ease of offering electrons to the unoccupied d orbital of metallic copper and the greater the inhibition efficiency [59]. E_{LUMO} indicates the ability of the molecules to accept electrons. The lower values of the E_{LUMO}, the more probable it is that the molecule would accept electrons. A low value of the energy bond gab ($\Delta E = E_{LUMO} - E_{HOMO}$) gives good inhibition efficiencies, because the energy to remove an electron from the last occupied orbital will be low [60]. Evaluation of the efficiency of ampicillin, amoxicillin, streptoquin and septazole as corrosion inhibitors for the copper in 0.1 M HCl solutions has been performed using MNDO method. The Mullikan charge densities, HOMO and LUMO electronic density distribution of ampicillin and amoxicillin were plotted in Figure 16. From the values of Mullikan charge we can observe the presence of excess of negative charge on nitrogen, oxygen and sulfur atoms can be adsorbed on the copper surface using these active centers leading to the corrosion inhibition action. Table 7 shows the quantum chemical calculation parameters for ampicillin and amoxicillin. From this tables, the smaller energy gap (ΔE) obtained in the case of amoxicillin can be interpreted by a stronger adsorption bond and perhaps greater inhibitor efficiency [61]. The less negative E_{HOMO} and the smaller ΔE reflect a great inhibition efficiency of the inhibitors [62]. From the energy of highest occupied molecular orbital (E_{HOMO}), the order of inhibiting efficiency is as follow: Amoxicillin > Ampicillin.

Mechanism of corrosion inhibition

The adsorption of investigated drug compounds can be attributed to the presence of polar unit having atoms of nitrogen, sulphur and oxygen and aromatic/heterocyclic rings. Therefore, the possible reaction centers are unshared electron pair of hetero-atoms and л-electrons of aromatic ring [63].

The adsorption and inhibition effect of investigated compounds in 0.1 M HCl solution can be explained as follows: In general, two modes of adsorption are considered on the metal surface in acid media. In the first mode, the neutral molecules may be adsorbed on the surface of copper through the chemisorption mechanism, involving the displacement of water molecules from the copper surface and the sharing electrons between the hetero- atoms and Cu.

The inhibitor molecules can also adsorb on the copper surface on the basis of donor–acceptor interactions between л - electrons of the aromatic ring and vacant d-orbitals of surface copper atoms. Thus we can conclude that inhibition of copper corrosion in 0.1 M HCl is mainly due to electrostatic interaction. The decrease in inhibition efficiency with rise in temperature supports electrostatic interaction. The order of inhibition is decreased as the following order: amoxicillin > ampicillin. This due to its larger molecular weight and the presence of OH group in it, which increases the electron charge density on the molecule.

Conclusions

The results obtained show that amoxicillin and ampicillin are good corrosion inhibitors for copper under acidic conditions. The maximum inhibition efficiency was 96%. Excellent agreement between the inhibition efficiencies calculated using different techniques was obtained. The adsorption of the amoxicillin and ampicillin onto the steel surface was characterized by the decrease in: (i) the cathodic and anodic current densities observed in the potentiodynamic polarization curves carried out in the presence of amoxicillin and ampicillin, (ii) the double-layer capacitance computed from electrochemical impedance spectroscopy experiments and (iii) electrochemical frequency modulation (EFM). The adsorption behavior of the amoxicillin and ampicillin is consistent with Langmuir adsorption isotherm. Amoxicillin and ampicillin are adsorbed on copper surface following physisorption and chemisorption mechanism. The results of polarization indicated that amoxicillin and ampicillin are of mixed type. A good agreement was obtained between all the investigated electrochemical techniques.

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