



## Investigating the Effect of Penicillin G as Environment-friendly Corrosion Inhibitor for Mild Steel in H<sub>3</sub>PO<sub>4</sub> Solution

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### ABSTRACT

*The corrosion behavior of mild steel (Ck 45) in 3.0 M H<sub>3</sub>PO<sub>4</sub> solution in the absence and presence of penicillin G was studied using potentiodynamic polarization and electrochemical impedance spectroscopy (EIS) techniques. The best inhibition effect at 10 mM of drug was a marked characteristic of the inhibitor. It was found that the inhibition efficiency of the inhibitor depends on its concentration and chemical structure. In other words, the inhibition efficiency increases with the inhibitor concentration. The adsorption process obeyed Langmuir adsorption isotherm. Some thermodynamic parameters were determined from the effect of temperature on corrosion and inhibition processes. Potentiodynamic polarization measurements indicated that the drug was a mixed type inhibitor. Prog. Color Colorants Coat. 11 (2018), 137-147 © Institute for Color Science and Technology.*

### 1. Introduction

Corrosion may be defined as an unintentional attack on a material through reaction with a surrounding medium. It is necessary to devote more attention to metallic corrosion nowadays, due to the growing usage of metals within different fields of technology and also the usage of rare and expensive metals for special applications, such as in atomic energy field. On the other hand, more corrosive environments due to the increasing of pollution of air and water is another reason for the interesting to the corrosion control in recent years.

A corrosion inhibitor is a substance which, when added to an environment in a small concentration, effectively reduces the corrosion rate of a metal exposed to that environment [1, 2]. However, because of numerous destructive effects that they have created in the environment, the use of these compounds as

inhibitor has been questioned recently, especially by environmentalists. Thus, the development of novel non-toxic or low-toxic corrosion inhibitors from natural sources has been considered as an important and desirable issue [3-8]. Recently studies have paid attention to the development of antibacterial drugs as environment-friendly inhibitors for metallic corrosion [6, 9-14]. Abdallah has described the inhibition effect of ampicillin, cloxacillin, flucloxacillin and amoxicillin in 2 M HCl solution by the formation of stable complex on the aluminium surface [9]. Golestani et al. have investigated the effect of penicillin G, ampicillin and amoxicillin on the corrosion behavior of mild steel in 1.0 M HCl solution [13]. The corrosion inhibition of mild steel in 1 M sulfuric acid using amoxicillin as inhibitor has been investigated by Kumar et al. [12].

In this work, the inhibition effect of penicillin G on corrosion of mild steel (Ck45) is investigated in 3 M

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$\text{H}_3\text{PO}_4$  solution. The main reason for choosing penicillin G as corrosion inhibitor was its low toxicity and high solubility in acidic media. The major disadvantage of using this drug as corrosion inhibitor is the risk of losing its effectiveness in remedial areas. When antibacterial drugs reach the wastewater and because of having long-lives they may lead to the development of resistant strains of bacteria [15]. The resistant bacteria may cause diseases that cannot be treated by the conventional antibacterial drugs. However, many different methods have been developed for degradation and removal of antibacterial drugs in aqueous solutions [16]. Adsorption, as a removal method, presents an efficient and economical technique widely used to accumulate antibacterial drugs from contaminated aqueous solutions onto a solid adsorbent [15-19]. The saturated adsorbent is easily separated from the treated solution for regeneration or disposal in an environmentally acceptable way. This paper has mainly an academic interest. The field application of the inhibitors requires further investigation [20]. If the further research confirms using of them in practice, a removal method must be employed before discharging the drugs solutions into the environment.

In this paper, the inhibition effect of penicillin G on corrosion of mild steel (CK 45) is investigated in 3 M  $\text{H}_3\text{PO}_4$  solution by using potentiodynamic polarization and electrochemical impedance spectroscopy (EIS) measurements.

## 2. Materials and Methods

### 2.1. Materials

Antibacterial drug, named penicillin G, was supplied from Sigma and used without further purification. The chemical structure of antibacterial drug is shown in Figure 1. The working electrode used here is made of mild steel (Ck45) with surface area of  $100 \text{ mm}^2$ .

### 2.2. Methods

EIS and potentiodynamic polarization measurements were used to investigate the corrosion rate of mild steel in 3 M  $\text{H}_3\text{PO}_4$  solution in the absence and presence of drug in various concentrations. Before carrying out the tests, the surface of the sample was rubbed with wet sandpapers through different grades then washed with distilled water and at last dried in air. The sample, as the working electrode (WE), was attached to a copper wire at one side, and other side was sealed by epoxy resin.

A potentiostat/galvanostat Autolab 302N (Eco Chemie, Netherlands) supported by a frequency response analyzer FRA-2 and Nova 1.9 software was used for performing Potentiodynamic polarization and EIS tests. The counter electrode (CE) was prepared from a rod of platinum with surface area of  $100 \text{ mm}^2$  and reference electrode was a saturated (KCl) Ag/AgCl electrode. The electrochemical order was EIS and afterward polarization technique. Before performing the tests, the specimens were immersed in the solution for about 30 min to obtain the stabilized open circuit potential (OCP).

A sinusoidal potential perturbation of 10 mV versus OCP in the 100 kHz-10 mHz frequency range was used in the EIS measurements. Nyquist plots of EIS data were analyzed by Nova 1.9 software.

Tafel curves were recorded at a scan rate of 1 mV/s and Nova 1.9 software was used to determine corrosion current density and polarization parameters. The samples in the absence and presence of 10 mM inhibitor were immersed for 24 h in 3 M  $\text{H}_3\text{PO}_4$  solution at room temperature. FT-IR spectra were recorded in a Bruker Tensor 27 spectrophotometer (Bruker, Ettlingen, Germany) which extended from 700 to  $4000 \text{ cm}^{-1}$  by using KBr disk method. For preparing FT-IR disk, small amount of drug was mixed with KBr and pressed.

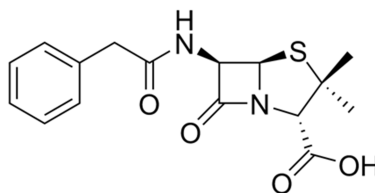


Figure 1: Structure of penicillin G.

Then the FT-IR spectra were obtained. The mild steel samples were rubbed by various sandpapers, washed with distilled water and dried at room temperature. After that, the samples were immersed in 3 M H<sub>3</sub>PO<sub>4</sub> solution for 24 h with specified amount of inhibitor, cleaned with distilled water and dried at room temperature. Afterward, the thin adsorbed layer on the mild steel surface was abraded by a small portion of KBr powder in an agate mortar to make a KBr disk.

### 3. Results and Discussion

#### 3.1. Potentiodynamic polarization

The Tafel curves of mild steel in 3 M H<sub>3</sub>PO<sub>4</sub> solution in the absence and presence of different concentrations of penicillin G are shown in Figure 2. The relevant parameters as corrosion current density (*i*<sub>corr</sub>), corrosion potential (*E*<sub>corr</sub>), anodic and cathodic Tafel slopes ( $\beta_a$ ,  $\beta_c$ ) are listed in Table 1. Corrosion current density decreased by increasing the concentration of inhibitor. Both cathodic and anodic branches of the potentiodynamic polarization curves are influenced by

the addition of inhibitor to H<sub>3</sub>PO<sub>4</sub> solution. Therefore, this drug behaved as a mixed inhibitor. However, corrosion potential shifted more noticeably to negative direction and this shows that the effect of inhibitor on the cathodic reaction is more visible than on the anodic reaction.

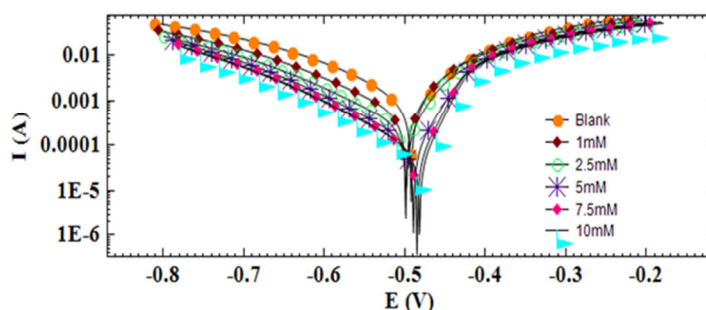
Table 1 shows the values of corrosion inhibition efficiency (IE) expressed by the following equation (Eq. 1) [20]:

$$IE_p(\%) = \frac{i_{corr} - i_{corr}^i}{i_{corr}} \times 100 \quad (1)$$

where *i*<sub>corr</sub> and *i*<sub>corr</sub><sup>i</sup> are corrosion current densities in the uninhibited and inhibited solutions, respectively. IE<sub>p</sub> values confirm that the inhibition of drug becomes more noticeable with increasing the inhibitors concentration. An increase in the inhibitor concentration up to 25 mM did not increase the IE (not included here). These values also indicate that the drug acts as an effective inhibitor.

**Table 1:** Polarization parameters and the corresponding inhibition efficiencies for mild steel in 0.5 M H<sub>3</sub>PO<sub>4</sub> containing different concentrations of inhibitor at 25 °C.

C /mM	<i>i</i> <sub>corr</sub> /μA.cm <sup>-2</sup>	- <i>E</i> <sub>corr</sub> /mV	$\beta_a$ /mV.decade <sup>-1</sup>	$\beta_c$ /mV.decade <sup>-1</sup>	IE <sub>p</sub> (%)
0	3435	493	172.9	235.9	-
1	1232	499	190.9	183.4	64
2.5	617	499	94.1	155.5	82
5	512	490	81.5	174.5	85
7.5	419	485	63.4	183.8	88
10	179	482	47.5	144.2	95



**Figure 2:** Polarization curves for mild steel in 3 M H<sub>3</sub>PO<sub>4</sub> solution in the absence and presence of different concentrations of penicillin G at 25 °C.

To calculate the surface coverage,  $\theta$ , it was assumed that the inhibition efficiency is due mainly to the blocking effect of the adsorbed species and hence  $\theta = \text{IE}(\%) / 100$ . An effort was made here to test the Langmuir, Temkin and Frumkin isotherms. The Langmuir adsorption isotherm was compatible with the experimental data (Figure 3), which can be expressed in following way (Eq. 2).

$$\frac{C}{\theta} = C + \frac{1}{K} \quad (2)$$

where  $\theta$  is the surface coverage,  $C$  is the inhibitor concentration and  $K$  is the adsorption equilibrium constant. As can be seen in Figure 3, the plot of  $C/\theta$  vs.

$C$  for the drug gives straight line with correlation coefficient close to 1 which confirms that the adsorption of this inhibitor is well described by the Langmuir adsorption isotherm.

### 3.2. Electrochemical impedance spectroscopy

Figure 4 shows Nyquist curves for mild steel in 3 M  $\text{H}_3\text{PO}_4$  in the absence and presence of different concentrations of penicillin G. Figure 4 obviously indicates that increasing the concentration of inhibitor has a considerable effect on the impedance response of the mild steel. Figure 5 shows the electrical equivalent circuit employed to analyze the impedance plots.

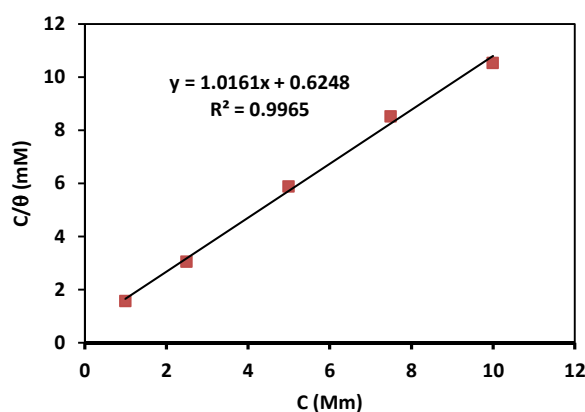


Figure 3: Langmuir adsorption isotherm of the inhibitor calculated by Tafel polarization results.

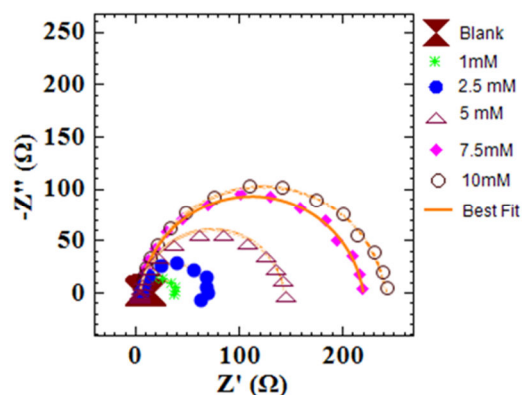


Figure 4: Nyquist plots for mild steel in 3 M  $\text{H}_3\text{PO}_4$  solution in the absence and presence of different concentrations of drug at 25°C.

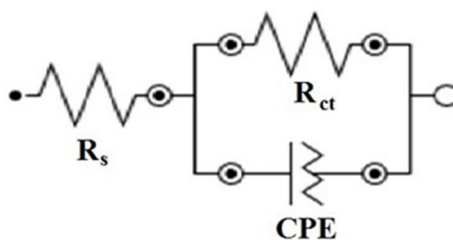


Figure 5: The equivalent circuit used to fit the experimental data.

EIS parameters in the absence and presence of different concentrations of penicillin G are listed in Table 2. It is evident that  $R_{ct}$  values increased by increasing the concentration of inhibitor. Besides, increasing the inhibitor concentration make a decrease in the values of  $C_{dl}$ . This condition is due to increasing the surface coverage by the inhibitor, which can lead to higher inhibition efficiency.

The values of IE in Table 2 were computed using the following equation (Eq. 3).

$$IE_{EIS}(\%) = \frac{R_{ct}' - R_{ct}}{R_{ct}'} \times 100 \quad (3)$$

where  $R_{ct}$  and  $R_{ct}'$  are the charge transfer resistance before and after addition of the inhibitor to the corrosive media, respectively. IE values increased as the concentration of inhibitor increased but there is a restriction in the sense that an additional increase in the inhibitor concentration did not make any considerable

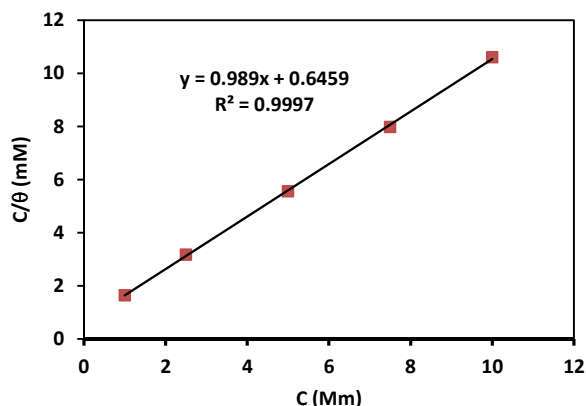
change in the inhibition efficiency of inhibitor. The IE values acquired from the EIS method (Table 2) are completely consistent with those acquired from the polarization method and actually confirm each other (Table 1). As can be seen from Figure 6, the EIS results are compatible with the Langmuir isotherm.

### 3.3. Temperature effect and thermodynamic parameters

Potentialdynamic polarization measurements in the range of 25-55 °C in the absence and presence of different concentrations of drug were used to investigate the adsorption of inhibitor and to determine the  $E_a$  and thermodynamic parameters of the mild steel corrosion process in acidic solution. The Tafel curves in the absence and presence of 10 mM drug are demonstrated in Figure 7. Corrosion factors at various temperatures are listed in Table 3.

**Table 2:** Impedance parameters and the corresponding inhibition efficiency values for mild steel in 3 M  $H_3PO_4$  containing different concentrations of penicillin G at 25 °C.

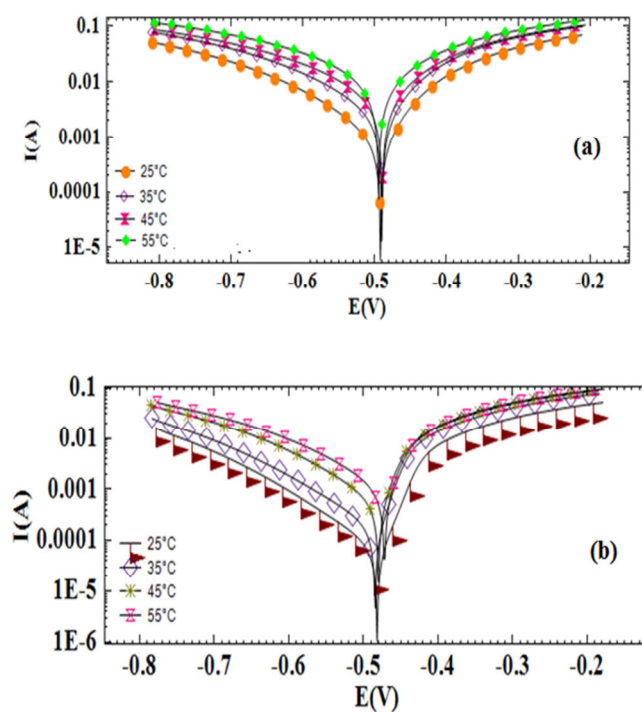
C/mM	$R_s/\Omega.cm^2$	$R_{ct}/\Omega.cm^2$	n	$C_{dl}/\mu F.cm^2$	$IE_{EIS}(\%)$
0	2.37	13.7	0.888	77.2	-
1	2.84	35.3	0.916	86.6	61
2.5	3.2	66.3	0.921	54.8	79
5	3	141	0.914	45.5	90
7.5	3.27	216	0.91	34.8	94
10	3.44	238	0.909	32.2	94.3



**Figure 6:** Langmuir adsorption isotherm of the inhibitor calculated by EIS results.

**Table 3:** Effect of temperature on the corrosion parameters of mild steel in 3.0 M H<sub>3</sub>PO<sub>4</sub> containing 0 and 10 mM penicillin G.

T/°C	C/mM	$i_{\text{corr}}/\mu\text{A}\cdot\text{cm}^{-2}$	IE%	$\theta$
25	0	3435	-	-
	10	179	94.8	0.948
35	0	7680	-	-
	10	1990	87.2	0.872
45	0	13410	-	-
	10	3830	74.8	0.748
55	0	15490	-	-
	10	5410	65.1	0.5

**Figure 7:** Effect of temperature on the polarization curves in 3 M H<sub>3</sub>PO<sub>4</sub> solution (a) without inhibitor and (b) in the presence of 10 mM of penicillin G.

The results acquired from the polarization curves proved an increase in  $i_{\text{corr}}$  and decrease in IE% by increasing temperature. In general, the reason for reducing the inhibition efficiency that occurs as a result of rising temperatures can be expressed as follows. The increase in temperature shortened the time lag between the adsorption and desorption of drug molecules on the surface of mild steel [21]. Therefore, the mild steel surface is longer exposed to the acidic media, resulting in an increase in corrosion rate with increasing the temperature, and then the values of IE% fall at high

temperatures.

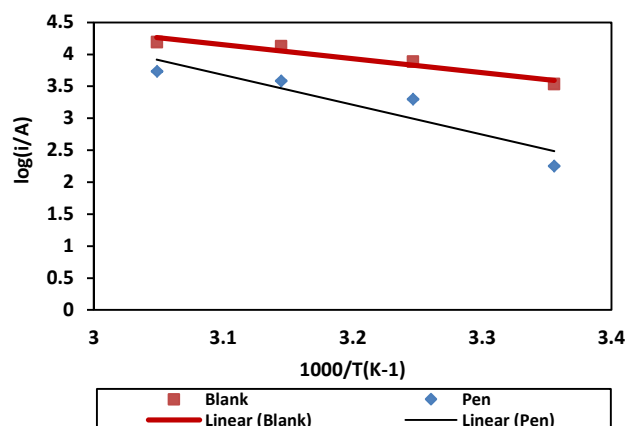
Arrhenius equation can express the dependence of the corrosion rate on temperature (Eq. 4).

$$i_{\text{corr}} = A \exp\left(\frac{-E_a}{RT}\right) \quad (4)$$

where  $i_{\text{corr}}$  is corrosion current, A is constant,  $E_a$  is the activation energy, R is the gas constant and T is the absolute temperature.  $E_a$  values can be obtained from the slope of Arrhenius plot [ $\log i_{\text{corr}}$  vs  $1/T$  (Figure 8)].

**Table 4:** Activation and thermodynamic parameters of adsorption obtained by potentiodynamic polarization measurements for mild steel in 3.0 M H<sub>3</sub>PO<sub>4</sub> solution in the absence and presence of 10 mM of penicillin G.

Drug	E <sub>a</sub> (kJ.mol <sup>-1</sup> )	K <sub>ads</sub> (M <sup>-1</sup> )	ΔG <sub>ads</sub> (kJ.mol <sup>-1</sup> )	ΔH <sub>ads</sub> (kJ.mol <sup>-1</sup> )	ΔS <sub>ads</sub> (J.K <sup>-1</sup> . mol <sup>-1</sup> )
Blank	18	-	-	-	-
Pen	41.1	1634	-28.3	-62.6	-115


**Figure 8:** Arrhenius plots for mild steel in 3 M H<sub>3</sub>PO<sub>4</sub> solution in the absence (blank) and presence of 10 mM inhibitor.

Computed E<sub>a</sub> values in the absence and presence of 10 mM penicillin G are shown in Table 4. The E<sub>a</sub> value acquired for the corrosion of mild steel in H<sub>3</sub>PO<sub>4</sub> solution (18.0 kJ.mol<sup>-1</sup>) is in a good agreement with those reported in other investigations [22-24]. Reducing the inhibition efficiency by increasing the temperature, with a similar increase in the corrosion activation energy in the presence of 10 mM inhibitor compared with its absence, is often interpreted as an offer to form an adsorption film from the physical (electrostatic) nature [25, 26].

The value of adsorption equilibrium constant, K<sub>ads</sub>, can be calculated by the reciprocal of the intercept of isotherm line (Figure 3). The free energy of inhibitor adsorption on mild steel surface is obtained from the following equation (Eq. 5).

$$\Delta G_{ads} = -RT \ln(55.5 K_{ads}) \quad (5)$$

where 55.5 is the molar concentration of water in the solution (in M), R is the gas constant (8.314 J K<sup>-1</sup>.mol<sup>-1</sup>) and T is the absolute temperature (K). The values of K<sub>ads</sub> and ΔG<sub>ads</sub> obtained from Tafel and EIS measurements are summarized in Table 5. An acceptable conformity was found between the two techniques.

The obtained ΔG<sub>ads</sub> value for mild steel in H<sub>3</sub>PO<sub>4</sub> solution was about -28 kJ/mol. Generally, if ΔG<sub>ads</sub> values are about -20 kJ/mol or less negative, it can be said that physical adsorption has occurred and if ΔG<sub>ads</sub> values are about -40 kJ mol<sup>-1</sup> or more negative kJ/mol it can be said that chemical adsorption has occurred. Therefore, it can be concluded from the obtained value for ΔG<sub>ads</sub> that the adsorption of mentioned drug is not only chemisorption or physisorption but involving comprehensive adsorption (both chemical and physical adsorption).

**Table 5:** The values of K<sub>ads</sub> and ΔG<sub>ads</sub> corresponding to polarization and EIS data in 3.0 M H<sub>3</sub>PO<sub>4</sub> solution.

Drug	Tafel		EIS	
	K <sub>ads</sub> (M <sup>-1</sup> )	ΔG <sub>ads</sub> (kJ.mol <sup>-1</sup> )	K <sub>ads</sub> (M <sup>-1</sup> )	ΔG <sub>ads</sub> (kJ.mol <sup>-1</sup> )
Pen	1634	-28.3	1586	-28.2

Langmuir adsorption isotherm is expressed by the following equation [27] (Eq. 6).

$$\ln\left(\frac{\theta}{1-\theta}\right) = \ln A + \ln C - \frac{\Delta H_{ads}}{RT} \quad (6)$$

where  $\theta$  is surface coverage,  $A$  is independent constant,  $C$  is concentration,  $R$  is gas constant,  $T$  is absolute temperature, and  $\Delta H_{ads}$  is enthalpy of adsorption.

The plot of  $\ln(\theta/(1-\theta))$  versus  $1/T$  at constant additive concentration gives a straight line as shown in Figure 9. The slope of the straight line is  $-\Delta H_{ads}/R$ . The calculated value of  $\Delta H_{ads}$  for inhibitor adsorption is given in Table 4. The negative values of  $\Delta H_{ads}$  represent the exothermic behavior of inhibitor on the mild steel surface. In an exothermic process, the absolute value of adsorption enthalpy can distinguish physisorption from chemisorption. In general, the values of  $\Delta H_{ads}$  around  $40 \text{ kJ mol}^{-1}$  or lower than are consistent with a physisorption process, and those around  $100 \text{ kJ mol}^{-1}$  indicate a chemisorptions process the absolute enthalpy of a physisorption process is lower than  $40 \text{ kJ.mol}^{-1}$  [28]. In this paper, the value of enthalpy, which is larger than the usual physical adsorption heat and is smaller than the chemical adsorption heat, it can be explained by the fact that the adsorption was carried out both physically and chemically.

Entropy of adsorption ( $\Delta S_{ads}$ ) can be calculated using the following equation (Eq. 7).

$$\Delta G_{ads} = \Delta H_{ads} - T\Delta S_{ads} \quad (7)$$

The obtained values for  $\Delta S_{ads}$  are shown in Table 4. The negative values of  $\Delta S_{ads}$  can be explained as follows. Inhibitor molecules before the adsorption over the mild steel surface may freely move in the bulk solution (the inhibitor molecules are disordered), but they are orderly adsorbed over the metal surface with the adsorption progression, and finally reduces the entropy (and caused a decrease in entropy) [29]. Hence, the negative value of  $\Delta S_{ads}$  in the present study emphasizes the high absorbability of the drug over the metal surface.

### 3.4. Fourier transform infrared (FT-IR) spectroscopy

The FT-IR spectra of pure penicillin G and the adsorbed layer formed on mild steel surface after 24 hr immersion in 3 M  $\text{H}_3\text{PO}_4$  containing 10 mM drug are shown in Figure 10.  $\beta$ -Lactams fused to thiazolidine rings show carbonyl absorption between  $1780$  and  $1770 \text{ cm}^{-1}$  [30]. Therefore, the strong bands at  $1780 \text{ cm}^{-1}$  can be attributed to stretching of carbonyl group of  $\beta$ -Lactam four-membered ring. The bands at  $1693 \text{ cm}^{-1}$  in penicillin G (Figure 10a) are assigned to C=O stretching of amide group. The absorption band around  $3300 \text{ cm}^{-1}$  to  $3400 \text{ cm}^{-1}$  is due to N-H stretching. The absorption bands near  $2960 \text{ cm}^{-1}$  are attributed to the aromatic C-H stretching vibrations.

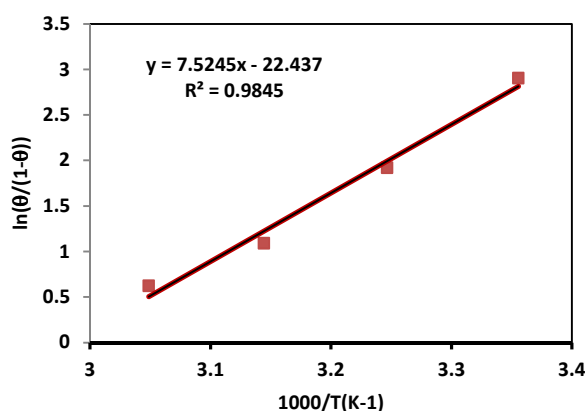
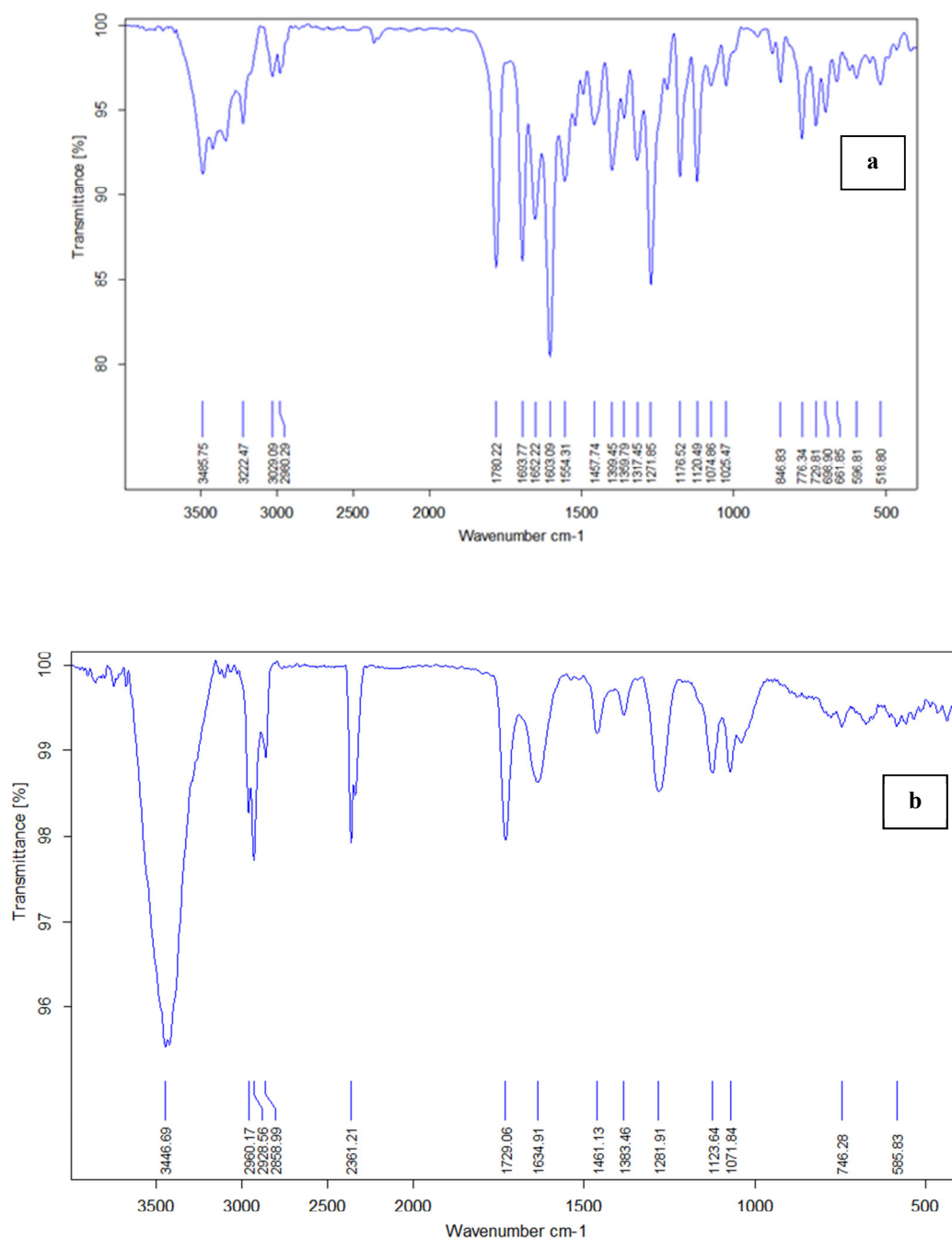


Figure 9: Plot of  $\ln(\theta/(1-\theta))$  vs.  $1/T$  for mild steel in 3 M  $\text{H}_3\text{PO}_4$  solution containing 10 mM of drug.





**Figure 10:** FT-IR spectra of (a) pure penicillin G and (b) adsorbed layer formed on mild steel after 24 h immersion in 3 M H<sub>3</sub>PO<sub>4</sub> solution + 10 mM of penicillin G.

By comparison, we found that the majority of the bands observed in the spectrum of adsorbed layer on mild steel (Figure 10b) are very similar to those appear in the pure drug spectrum (Figure 10a). The bands at 2337 and 2361 cm<sup>-1</sup> (Figure 10a) are allocated to CO<sub>2</sub> bands [31, 32].

A broad band at around 3440 cm<sup>-1</sup> is attributed to

O-H stretching, which further indicates that the adsorbed film contains H<sub>2</sub>O. The weak bands near 2960 cm<sup>-1</sup> are known as aromatic C-H stretching vibrations. The disappearance of the C=O stretching vibration (1780 cm<sup>-1</sup>) indicates that oxygen atom of β-lactam ring can act as an active agent in adsorption. The disappearing of C=O stretching band of amide

group in Figure 10b also indicates that oxygen atom of amide group is an active center in adsorption. Therefore, it can be concluded that the drug is actually adsorbed on the surface of the mild steel.

#### 4. Conclusions

The adsorption and inhibition effects of an antibacterial drug, named penicillin G, on the corrosion behavior of mild steel in 3.0 M H<sub>3</sub>PO<sub>4</sub> were studied using electrochemical techniques such as Tafel and EIS. The

data acquired from potentiodynamic polarization and EIS measurements indicated that the adsorption of penicillin G on mild steel in 3.0 M H<sub>3</sub>PO<sub>4</sub> follows the Langmuir isotherm. The calculated values for the free energy and enthalpy of adsorption showed that both physical and chemical adsorption may occur, but it seems that the adsorption of the studied drug is mainly chemisorption. The IE and  $\Delta G_{\text{ads}}$  values obtained from Tafel method showed an acceptable compatibility with the data acquired from EIS technique.

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