



Investigation of adsorption and corrosion inhibition of mild steel in hydrochloric acid solution by 6-phenylpyridazin-3(2H)-one

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ABSTRACT

In this study, the inhibition effect of 6-phenylpyridazin-3(2H)-one (GPO) on mild steel corrosion in 1.0M HCl solution was studied. For this aim, electrochemical techniques such as potentiodynamic polarization curves, weight loss (WL) and electrochemical impedance spectroscopy (EIS) were used. It was shown that, the GPO base has remarkable inhibition efficiency on the corrosion of mild steel in 1.0M HCl solution. Polarization measurements indicated that, the studied inhibitor acts as mixed type corrosion inhibitor with predominantly control of cathodic reaction. The inhibition efficiency depends on the concentration of inhibitor and reaches 82.7% at 1.0mM GPO. The remarkable inhibition efficiency of GPO was discussed in terms of blocking of electrode surface by adsorption of inhibitor molecules through active centers. The adsorption of GPO molecules on the mild steel surface obeys Langmuir adsorption isotherm.

Keywords: Mild Steel, Acid Corrosion, Inhibitor, Electrochemical techniques, Gravimetric.

INTRODUCTION

Acid solutions are generally used for removal of undesirable scale and rust on the metals, cleaning of boilers and heat exchangers, oil-well acidizing in oil recovery, and so on. [1-3]. HCl solution is one of the most widely used agents for these goals. However, iron and its alloys could be corroded during these applications which result in a waste of resources. Corrosion prevention systems favor the use of chemicals with low or no environmental impacts. The reduction in the corrosion rate of metals has numerous advantages such as saving of resources, economic benefits during the industrial applications, increasing the lifetime of equipment and decreasing the dissolution of toxic metals from the components into the environment. Therefore, the prevention of metals against corrosion is vital and must be dealt with.

The existing data show that organic inhibitors act by adsorption and a film formation on the surface of metals. The adsorption of organic inhibitors on the metal surface can change the corrosion resistance properties of metals. Earlier studies have shown that organic compounds bearing heteroatoms with high electron density such as phosphorus, sulfur, nitrogen, oxygen or those containing multiple bonds, which are considered as adsorption centers, are effective inhibitors for the corrosion of metals [4-31]. The organic inhibitors are generally adsorbed on the metal surface through physical adsorption or chemical adsorption, which reduce the reaction area susceptible to corrosive attack [32,33].

The aim of this study is to investigate inhibition effect of 6-phenylpyridazin-3(2H)-one (GP0) GP0 on the mild steel corrosion in 1.0 M HCl solution. For this purpose, potentiodynamic polarization, WL and EIS techniques were used. The chemical structure of the studied GP0 derivative is given in Fig 1.

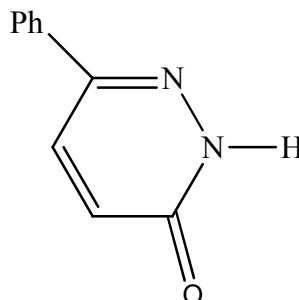


Figure 1. The chemical structure of the studied pyridazine derivative compound

EXPERIMENTAL SECTION

Materials

The steel used in this study is a mild steel (MS) with a chemical composition (in wt%) of 0.21 % C, 0.38 % Si, 0.05 % Mn, 0.05 % S, 0.09 % P, 0.01 % Al and the remainder iron (Fe).

Solutions

The aggressive solutions of 1.0 M HCl were prepared by dilution of analytical grade 37% HCl with distilled water. The organic compound tested is 6-phenylpyridazin-3(2H)-one (GP0). The concentration range of this compound was 10^{-3} to 10^{-6} M.

Weight loss measurements

The mild steel (MS) sheets of $1 \times 1 \times 0.05$ cm were abraded with a series of emery papers SiC (120, 600 and 1200) and then washed with distilled water and acetone. After weighing accurately, the specimens were immersed in an 80 mL beaker containing 50 mL

1.0 M HCl solution with and without addition of different concentrations of GP0. All the aggressive acid solutions were open to air. After 6 h the specimens were taken out, washed, dried, and weighed accurately. In order to get good reproducibility experiments were carried out in triplicate. The average weight loss of three parallel MS sheets was obtained. The tests were repeated at 308 K. The corrosion rate (v) and the inhibition efficiency (η_{WL}) were calculated by the following equations [34]:

$$v = \frac{W}{St} \times 100 \quad (1)$$

$$\eta_{WL} (\%) = \frac{v_0 - v}{v_0} \times 100 \quad (2)$$

where W is the three-experiment average weight loss of the mild steel, S is the total surface area of the specimen, t is the immersion time and v_0 and v are values of the corrosion rate without and with addition of the inhibitor, respectively.

Polarization measurements

Electrochemical impedance spectroscopy

The electrochemical measurements were carried out using Volta lab (Tacussel- Radiometer PGZ 100) potentiostat and controlled by Tacussel corrosion analysis software model (Voltmaster 4) at under static condition. The corrosion cell used had three electrodes. The reference electrode was a saturated calomel electrode (SCE). A platinum electrode was used as auxiliary electrode of surface area of 1 cm^2 . The working electrode was mild steel. All potentials given in this study were referred to this reference electrode. The working electrode was immersed in test solution for 30 minutes to a establish steady state open circuit potential (E_{ocp}). After measuring the E_{ocp} , the electrochemical measurements were performed. All electrochemical tests have been performed in aerated solutions at 308 K. The EIS experiments were conducted in the frequency range with high limit of 100 kHz and different low limit

0.1 Hz at open circuit potential, with 10 points per decade, at the rest potential, after 30 min of acid immersion, by applying 10 mV ac voltage peak-to-peak. Nyquist plots were made from these experiments. The best semicircle can be fit through the data points in the Nyquist plot using a non-linear least square fit so as to give the intersections with the x -axis.

The inhibition efficiency of the inhibitor was calculated from the charge transfer resistance values using the following equation [35]:

$$\eta_z \% = \frac{R_{ct(inh)} - R_{ct}}{R_{ct(inh)}} \times 100 \quad (3)$$

where R_{ct} and $R_{ct(inh)}$ were the values of polarization resistance in the absence and presence of inhibitor, respectively.

Potentiodynamic polarization

The electrochemical behaviour of mild steel sample in inhibited and uninhibited solution was studied by recording anodic and cathodic potentiodynamic polarization curves. Measurements were performed in the 1.0 M HCl solution containing different concentrations of the tested inhibitor by changing the electrode potential automatically from -800 to 200 mV versus corrosion potential at a scan rate of 1 mV s⁻¹. The linear Tafel segments of anodic and cathodic curves were extrapolated to corrosion potential to obtain corrosion current densities (I_{corr}). From the polarization curves obtained, the corrosion current (I_{corr}) was calculated by curve fitting using the equation:

$$I = I_{corr} \left[\exp\left(\frac{2.3\Delta E}{\beta_a}\right) - \exp\left(\frac{2.3\Delta E}{\beta_c}\right) \right] \quad (4)$$

The inhibition efficiency was evaluated from the measured I_{corr} values using the relationship:

$$\eta_{Tafel} \% = \frac{I_{corr}^{\circ} - I_{corr}^i}{I_{corr}^{\circ}} \times 100 \quad (5)$$

where, I_{corr}° and I_{corr}^i are the corrosion current density in absence and presence of inhibitor, respectively.

RESULTS AND DISCUSSION

Weight loss measurements

The weight loss of mild steel specimens after exposure to 1.0 M HCl solution with and without addition of various concentrations of GP0 was calculated and the data obtained were given in Table 1. It is clear from Table 1 that, the addition of inhibitor to the aggressive solution reduces dissolution rate of mild steel efficiently. The weight loss was reduced with increasing pyridazine derivative concentration indicates that the inhibitor molecules act by adsorption on the metal surface.

Table 1. Corrosion parameters for mild steel in aqueous solution of 1.0 M HCl in the absence and presence of different concentrations of inhibitor from weight loss measurements at 308K

Inhibitor	Conc (M)	v (mg/cm ² h)	η_{WL} (%)	θ
HCl	1.0	1.142	----	----
GP0	1×10^{-3}	0.197	82.7	0.827
	5×10^{-4}	0.200	82.4	0.824
	1×10^{-4}	0.345	69.8	0.698
	5×10^{-5}	0.438	61.5	0.615
	1×10^{-5}	0.525	54.0	0.540
	1×10^{-6}	0.696	38.9	0.389

Electrochemical Impedance Spectroscopy (EIS)

Electrochemical impedance spectroscopy (EIS) is an effective method for corrosion studies of metallic materials. The effect of GP0 concentration on the impedance spectra of mild steel in 1.0 M HCl solutions at 308 K is recorded in Fig. 2 (Nyquist plots). It is clear to see that the impedance spectra are significantly changed with addition of different GP0 concentration. From the Nyquist plots, it was also observed that, even the presence of GP0 does not

alter the style of impedance plots, thus indicating the addition of GP0 does not change the mechanism for the dissolution of mild steel in 1.0 M HCl solution [36,37].

The impedance diagrams show only one capacitive loop represented by slightly depressed semicircle which indicates that the corrosion of mild steel in 1.0 M HCl solution is mainly controlled by charge transfer process and formation of a protective layer on the mild steel surface. The diameter of the capacitive loop increases with the increase of GP0 concentration proposing that the formed inhibitive film was strengthened by the addition of GP0 [38]. The depressed semicircles are generally attributed to the frequency dispersion as well as roughness and inhomogeneities of solid surface, and mass transport process [39], distribution of the active sites, adsorption of inhibitors [40,41].

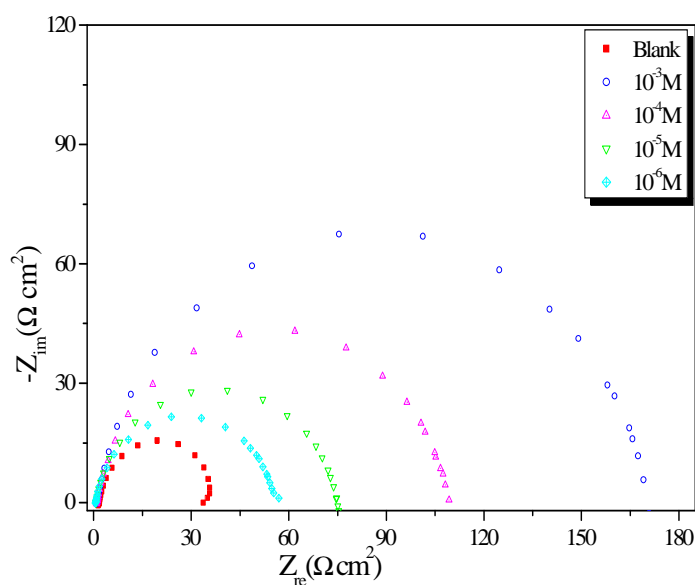


Figure 2. Nyquist diagrams for mild steel in 1.0 M HCl containing different concentrations of GP0 at 308K

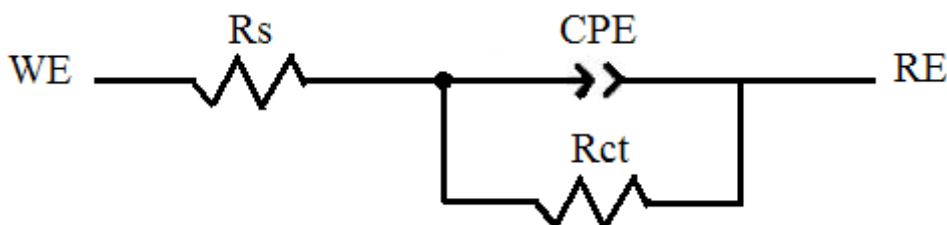


Figure 3. The electrochemical equivalent circuit used to fit the impedance measurements

Therefore, a constant phase element *CPE* is used to replace a capacitive element to acquire a more accurate fit of experimental data. Fig. 3 displays the equivalent circuit, which consists of a solution resistance R_s , the constant phase element *CPE* and the charge transfer resistance R_{ct} . The impedance of *CPE* is described as the mathematical expression given below [42]

$$Z_{CPE} = \frac{1}{Y_0} \times \frac{1}{(j\omega)^n} \quad (5)$$

Where Y_0 is the magnitude of the *CPE*, j is the imaginary unit, ω is the angular frequency and n is the phase shift.

The fitted impedance parameters derived from Nyquist diagrams and inhibition efficiency are listed in Table 2. Data in Table 2 shows that additional GP0 inhibits the corrosion of mild steel in 1.0 M HCl solution. By increasing the inhibitor concentration R_{ct} values and inhibition efficiency increase. The increase in the charge transfer resistance values with increasing inhibitor concentration suggests the formation of a protective layer on the mild steel electrode surface. This protective layer behaves as a hindrance for mass and charge transfer [43].

The inhibitive efficiencies calculated from EIS results show the same trend as those obtained from weight loss measurements.

Table 2. Corrosion parameters obtained by impedance measurements for mild steel in 1.0 M HCl at various concentrations of GP0

Inhibitor	Conc (M)	R_s ($\Omega \text{ cm}^2$)	R_{ct} ($\Omega \text{ cm}^2$)	f_{max} (Hz)	C_{dl} ($\mu\text{F}/\text{cm}^2$)	η_z (%)
HCl	1.0	1.68	33.23	50.00	95.8	-----
GP0	10^{-3}	1.30	169.84	17.86	52.5	80.4
	10^{-4}	0.92	108.23	22.32	65.9	69.3
	10^{-5}	0.78	074.65	28.09	75.9	55.5
	10^{-6}	1.20	054.09	31.65	93.0	38.6

Potentiodynamic polarization

Figure 4 illustrates the polarization curves of mild steel in 1.0 M HCl solution without and with various concentrations of GP0 at 308 K. The presence of GP0 shifts both anodic and cathodic branches to the lower values of corrosion current densities and thus causes a remarkable decrease in the corrosion rate. The parameters derived from the polarization curves in Figure 4 are given in Table 3. In 1.0 M HCl solution, the presence of GP0 causes a remarkable decrease in the corrosion rate i.e., shifts both anodic and cathodic curves to lower current densities. In other words, both cathodic and anodic reactions of mild steel electrode are retarded by GP0 in hydrochloric acid solution. The Tafel slopes of β_c at 308 K do not change remarkably upon addition of GP0, which indicates that the presence of GP0 does not change the mechanism of hydrogen evolution and the metal dissolution process. Generally, an inhibitor can be classified as cathodic or anodic type if the shift of corrosion potential in the presence of the inhibitor is more than 85 mV with respect to that in the absence of the inhibitor [44,45]. In the presence of GP0, E_{corr} shifts to less negative but this shift is very small (about 10-30 mV), which indicates that GP0 can be arranged as a mixed-type inhibitor, with predominant cathodic effectiveness.

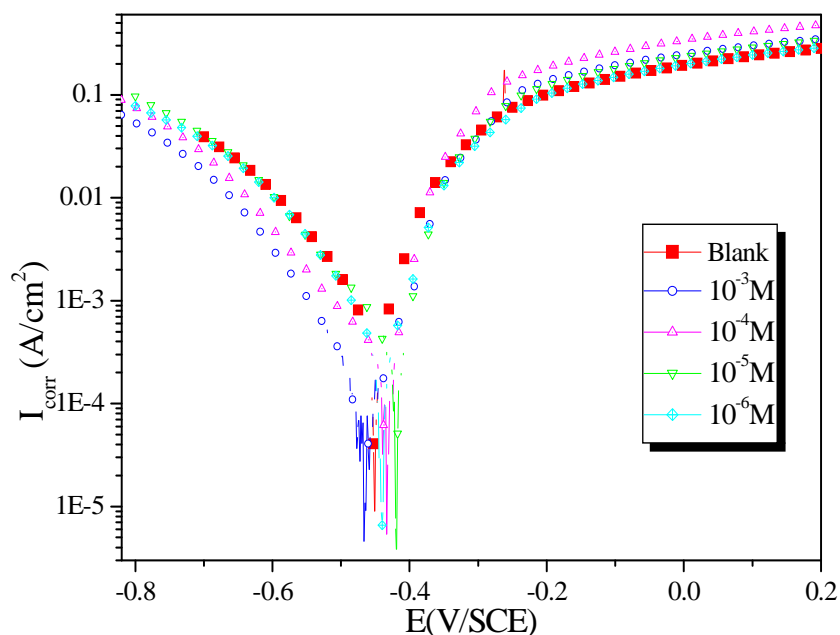


Figure 4. Potentiodynamic polarization curves for the corrosion of mild steel in 1.0 M HCl solution without and with various concentrations of GP0 at 308 K

Table 3. Effect of concentration of GP0 on the electrochemical parameters calculated using potentiodynamic polarization technique for the corrosion of mild steel in 1.0 M HCl at 308 K

Inhibitor	Conc (M)	-E _{corr} (mV/SCE)	-β _c (mV/dec)	I _{corr} (μA/cm ²)	η _{Tafel} (%)
HCl	1.0	455.2	127.3	815.7	-----
	10 ⁻³	469.8	118.7	159.8	80.4
	10 ⁻⁴	437.8	132.0	259.4	68.2
GP0	10 ⁻⁵	424.2	123.1	369.6	54.7
	10 ⁻⁶	445.3	116.8	495.0	39.3

Adsorption considerations

The mechanism of corrosion inhibition may be illustrated on the basis of adsorption isotherm. In order to obtain the adsorption isotherm, the degree of surface coverage (θ) of GP0 must be calculated. In this study, the degree of surface coverage values (θ) for various concentrations of GP0 in 1.0 M HCl solution have been obtained from Table 1. Attempts were made to fit the data to different isotherms, including Langmuir, Bockris-Swinkles, Temkin, Flory-Huggins and Frumkin. Among the isotherms, the best fit is the Langmuir isotherm. According to Langmuir adsorption isotherm, the relation of inhibitor concentration(C) and surface coverage (θ) is described by equation (6) [46]:

$$\frac{C}{\theta} = \frac{1}{K_{ads}} + C \quad (6)$$

Where C is the inhibitor concentration (mol L⁻¹), θ is the surface coverage, K_{ads} is the adsorption equilibrium constant (L mol⁻¹). K_{ads} is related to the Gibbs free energy of adsorption, ΔG_{ads}° (kJ mol⁻¹) as [47]:

$$K_{ads} = \frac{1}{C_{solvent}} \exp\left(\frac{-\Delta G_{ads}^{\circ}}{RT}\right) \quad (7)$$

where R (8.314 J mol⁻¹ K⁻¹) is the molar gas constant, T (K) is the absolute temperature, and $C_{solvent}$ is the concentration of the solvent, which in this case is water ($C_{H_2O} = 55.5$ mol L⁻¹).

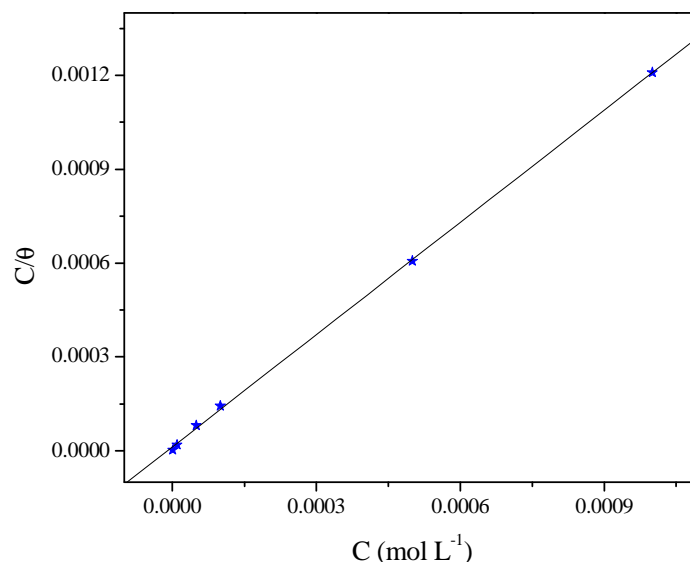
**Figure 5. Langmuir adsorption isotherm for mild steel in 1.0 M HCl containing different concentrations of GP0 at 308 K**

Fig.5 shows the linear plots of C/θ versus C were obtained at 308 K indicating that the adsorption of GP0 obeyed Langmuir isotherm. The various adsorption parameters obtained by fitting method from this isotherm are given in Table 4. It is seen from the Table 4 that the correlation coefficient ($r = 0.99984$) demonstrates that adsorption of GP0 on mild steel follows this isotherm and supposes that the adsorbed molecules occupy only one site and there are no

interactions with other adsorbed species [48]. According to Eq. (6), K_{ads} can be calculated from intercept of the plot of C/θ vs. C . With the above equation (7), ΔG_{ads}° can be calculated from K_{ads} and listed in Table 4 [48-50].

The average value of K_{ads} is $80546.4 \text{ L mol}^{-1}$, which reflects the high adsorption ability of GP0 on the mild steel surface. Namely, the adsorption process is more favorable than the desorption [51,52]. The ΔG_{ads}° is negative and high ($-39.2 \text{ kJ mol}^{-1}$) indicates the strong interactions between the inhibitor molecules and the metal surface [53-56]. Generally, the standard free energy of adsorption values of -20 kJ mol^{-1} or less negative are associated with physical adsorption; those of -40 kJ mol^{-1} or more negative involves charge sharing or transfer between inhibitor molecules and metals (chemical adsorption) [57-60]. However, the adsorption of organic molecules on metal surfaces cannot be considered as purely physical or chemical phenomenon. In addition to the chemical adsorption, inhibitor molecules can also be adsorbed on metal surface via physical interactions. In this study, the ΔG_{ads}° is modestly closer to -40 kJ mol^{-1} . Therefore, it is concluded that chemical interactions should be dominant for the adsorption of the GP0 molecules on the mild steel surface [61-63].

Table 4. Some parameters from Langmuir model for mild steel in 1.0 M HCl at 308K

Inhibitor	Slope	$K_{ads} (\text{M}^{-1})$	r	$\Delta G_{ads}^{\circ} (\text{kJ/mol})$
GP0	1.15	80596.41	0.99984	-39.21

CONCLUSION

The corrosion inhibition of mild steel in 1.0 M HCl solution by GP0 was studied using common electrochemical techniques and weight loss. According to experimental findings, it could be concluded that:

1. GP0 is a good corrosion inhibitor for the mild steel protection in 1.0 M HCl solution. The inhibitory efficiency of this compound depends on its concentration.
2. GP0 acts by reducing the rates of both anodic and cathodic reactions.
3. AC impedance results were interpreted using an equivalent circuit in which a constant phase element (CPE) was used in place of a double layer capacitance (C_{dl}) in order to give more accurate fit to the experimental results.
4. The high inhibition efficiency of the inhibitor was explained by adsorption of the GP0 molecules on the steel surface and a protective film formation.
5. The adsorption of GP0 on the steel surface from 1.0 M HCl solution follows Langmuir adsorption isotherm. The thermodynamic parameters suggest that this inhibitor is strongly adsorbed on the mild steel surface.

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