

Research Article



Study of 5-Chlorobenzodiazol-4-Piperidin-N-Benzodiazol Propane as Corrosion Inhibitor for Carbon Steel in 1M HCl Solution

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ABSTRACT

(Domperidone) 5-Chlorobenzodiazol-4-Piperidin-N-Benzodiazol Propane, (CPBP) as drugs was used as corrosion inhibitors for carbon steel. The inhibition efficiency of CPBP as corrosion inhibitor of carbon steel in 1 M HCl solution was studied by weight loss, electrochemical impedance spectroscopy (EIS) and potentiodynamic polarization techniques. The Results shows that the, inhibition efficiency increased with increasing of inhibitor concentrations and CPBP was used is a good inhibitor. The adsorption of Domperidone obeyed Langmuir adsorption isotherm. Both thermodynamic and activation parameters were calculated and discussed. Polarization curves indicated that the inhibitor act as mixed type. All the results obtained from different experimental studies are in a good agreement.

Keywords: Corrosion, Carbon Steel, Weight Loss, EIS, SEM

INTRODUCTION

Organic inhibitors were applied extensively to protect metals from corrosion in many aggressive acidic media e.g., in the acid pickling and cleaning processes of metals¹⁻³. Organic compounds containing N, S and O atoms⁴⁻¹² were found to be good corrosion inhibitors of metals particularly for active metals like Fe, Zn, Mg and Cu etc. The effectiveness of these compounds as corrosion inhibitors has been interpreted in terms of their molecular structure, molecular size, and molecular mass, heteroatom present and adsorptive tendencies¹³. Under certain conditions, the electronic structure of the organic inhibitors has a key influence on the corrosion inhibition efficiency to the metal. The inhibitors influence the kinetics of the electrochemical reactions which constitute the corrosion process and thereby modify the metal dissolution in acids. The existing data show that most organic inhibitors act by adsorption on the metal surface. Quite a number of studies have been carried out in determination of adsorptive of various compounds at the electrode/solution interface¹⁴⁻¹⁶.

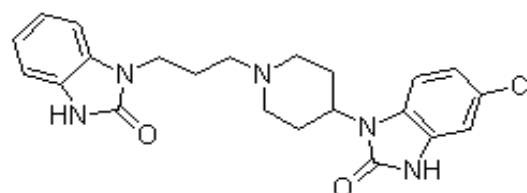
A large number of organic compounds were studied as corrosion inhibitor, unfortunately most of the organic inhibitors used are very expensive and health hazards. Their toxic properties limit their field of application. Thus, it remains an important object to find cost-effective and non-hazardous inhibitors for the protection of metals against corrosion. In this connection, the influences of nontoxic organic compounds and drugs on the corrosion of metals in acid media were investigated by several authors¹⁷⁻²⁵.

The objective of this work is to study the corrosion inhibitive action of Domperidone for the corrosion of carbon steel in 1M HCl. The choice of this drug as a corrosion inhibitor is based on its environmentally

friendly; its molecule has O, N atoms as active center atoms.

MATERIALS AND METHODS

Experimental



Chemical structure of 5chlorobenzodiazol-4-piperidin-N-benzodiazol propane (Domperidone)

Chemical Composition of Carbon Steel Sample

The experiments were performed with C-steel alloy having the following chemical composition. The weight percentages of Si, Sn, Mo, Zn, Cu, Ni, Mn, Cr, V, Ti, Al, S, C and Fe of the C-steel alloy are 0.913, 0.019, 0.103, 0.02, 0.135, 0.242, 0.955, 1.09, 0.078, 0.073, 0.570, 0.389, 0.133 and 95.28 respectively.

Electrochemical Techniques

Potentiodynamic Polarization

The electrical circuit used for determining the variation of electrode potential with the electrical current. The potentiodynamic current-potential curves were recorded by changing the electrode potential automatically with a scan rate 2 mV s⁻¹ from a low potential of -800 to -300 mV (SCE). Before each run, the working electrode was immersed in the test solution for 30 min to reach steady state. All potentials were measured against SCE. Measurements were obtained using a Voltalab 40 Potentiostat PGZ 301 combined with easy corrosion program (Voltmaster 4).



Electrochemical Impedance Spectroscopy (EIS)

Electrochemical impedance were obtained using a Voltalab 40 for all EIS measurements with a frequency range of 100 kHz to 50 mHz with a 4 mV sine wave as the excitation signal at open circuit potential. If the real part is plotted on the X-axis and the imaginary part is plotted on the Y-axis of a chart, we get a Nyquist Plot.

The charge transfer resistance values (R_{ct}) was calculated from the difference in impedance at lower and higher frequencies.

Weight Loss Techniques

The test coupons were cut into $2 \times 2 \times 0.2 \text{ cm}^3$. The samples were first mechanically polished with a fine grade emery paper in order to obtain a smooth surface, followed by degreasing in acetone⁶ then rinsed in distilled water and finally dried between two filter papers and weighed.

All experimental were carried out at 25, 35, 45 and 55 ± 1 °C.

RESULTS AND DISCUSSION

Electrochemical Studies

Potentiodynamic Polarization Techniques

The extrapolation of anodic and/or cathodic Tafel lines of charge transfer controlled corrosion reaction gives the corrosion current density, I_{corr} at corrosion potential, E_{corr} .

The potential (E) is plotted as a function of logarithm of current density (I) show the polarization curves for both anodic and cathodic reactions²⁶.

Figure 1 represent the potentiodynamic polarization plots for carbon steel electrode in 1M HCl in the absence and presence of different concentrations of the used inhibitor, at scanning rate 2mV/sec.

The percentage inhibition efficiency P % is given by:

$$P \% = \frac{(I_{corr} - I_{corr(inh)})}{I_{corr}} \times 100 \quad (1)$$

Where, I_{corr} and $I_{corr(inh)}$ are the corrosion current densities in absence and presence of inhibitors, respectively, determined by extrapolation of cathodic Tafel lines to the corrosion potential.

The degree of surface coverage (θ) was calculated the following equation:

$$\theta = \frac{I_{corr} - I_{corr(inh)}}{I_{corr}} \quad (2)$$

Table 1 shows the effect of inhibitor concentration on some electrochemical parameters, corrosion current density (I_{corr}), corrosion potential (E_{corr}), anodic and cathodic Tafel slopes (β_a and β_c), the degree of surface coverage (θ) and the percentage of inhibition efficiency (P %) during the corrosion of C-steel electrode in 1M HCl solutions.

Table 1: Potentiodynamic polarization parameters for corrosion of carbon steel in 1M HCl in absence and presence of different concentrations of CPBP at 25 °C at scanning rate 2mV s^{-1}

Conc. of inhibitor	E_{corr} mV	I_{corr} mA cm^{-2}	β_a mV dec^{-1}	β_c mV dec^{-1}	θ	P %
Blank	-542.2	0.4622	154.9	-319.6	---	---
100	-515.8	0.0638	107.6	-137.1	0.8619	86.19
200	-533.3	0.0598	281.3	-143.0	0.8706	87.06
300	-536.4	0.0581	122.6	-142.7	0.8742	87.42
400	-548.4	0.0555	218.6	-141.3	0.8799	87.99
500	-522.6	0.0362	118.7	-123.6	0.9267	92.67

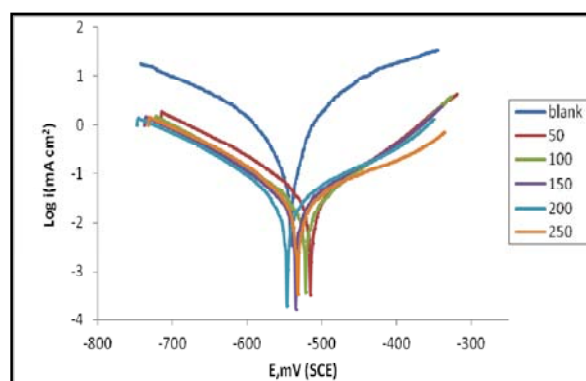


Figure 1: Potentiodynamic polarization curves for the carbon steel in 1M HCl in the absence and presence of different concentrations of CPBP at scanning rate 2mV s^{-1}

Electrochemical Impedance Spectroscopy (EIS)

The corrosion behavior of C-steel in 1M HCl solution in the absence and presence of the studied inhibitor was investigated by EIS method at 25 °C. Figure 2 shows the Nyquist plots for carbon steel in 1M HCl solution in the absence and presence of different concentrations of inhibitor at 25 °C. The Nyquist plots were regarded as one part of a semicircle.

The impedance diagram shows the same trend (one capacitive loop); however, the diameter of this capacitive loop increases with increasing concentration.

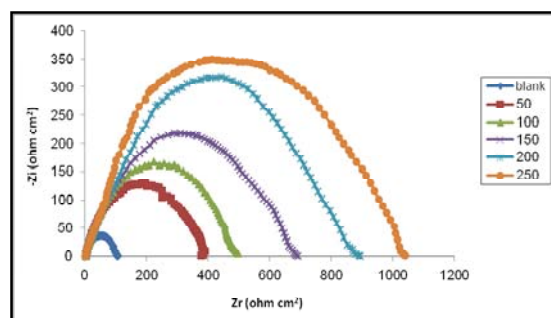


Figure 2: Nyquist plots for the carbon steel in 1M HCl in the absence and presence of different concentrations of CPBP

The presence of the inhibitor increases the impedance but does not change other aspects of the behavior.

These results support the results of polarization measurements that the inhibitor does not alter the electrochemical reactions responsible for corrosion. It inhibits corrosion primarily through its adsorption on the metal surface^{27,28}.

The charge transfer resistance values (R_{ct}) are calculated from the difference in impedance at lower and higher frequencies.

To obtain the double-layer capacitance (C_{dl}), the frequency at which the imaginary component of the impedance at maximum $f(-Z''_{img})$ is found and C_{dl} values are calculated from the following equation²⁶:

$$f(-Z''_{img}) = \frac{1}{(2\pi C_{dl} R_{ct})} \quad (3)$$

The impedance quantitative results can be seen in Table 2. It is clear that, the corrosion of steel is obviously inhibited in the presence of the inhibitor. It is apparent that, the impedance response for carbon steel in 1M HCl

Table 2: EIS parameters for corrosion of carbon steel in 1M HCl in the absence and presence of different concentrations of CPBP at 25 °C

Conc. of inhibitor	R_{ct} Ohm cm^2	R_s Ohm cm^2	C_{dl} μF cm^2	θ	P%
Blank	104.269	2.18	40.95	---	---
100	376.983	2.23	3.26	0.7234	72.34
200	496.703	1.51	1.92	0.7900	79.0
300	691.231	1.74	1.05	0.8491	84.91
400	894.012	1.76	0.56	0.8833	88.33
500	1040.325	1.94	0.43	0.8997	89.97

Table 3: Effect of CPBP inhibitor concentration on carbon steel corrosion in 1M HCl at 25 °C

Sample No.	Conc., ppm	Weight loss, mg/ cm^2	Corr. Rate, mg/ cm^2 .hr	θ	P (%)
Blank	0.0	0.48	0.69444	0.0	0.0
CPBP	100	0.1282	0.18547	0.7329	73.3
	200	0.1021	0.14771	0.7873	78.7
	300	0.0719	0.10402	0.8502	85.0
	400	0.0531	0.07682	0.8894	88.9
	500	0.0376	0.0544	0.9217	92.2

Table 4: Effect of temperature on carbon steel corrosion in presence of 500 ppm of CPBP inhibitor concentration in 1M HCl

Temp., °C	Weight loss, mg/ cm^2	Corr. Rate, mg/ cm^2 .hr	θ	P (%)
25	0.0165	0.0544	0.9217	92.2
35	0.0407	0.17665	0.8664	86.6
45	0.0927	0.40234	0.8279	82.8
55	0.1969	0.8546	0.7867	78.7

changes significantly with increasing inhibitor concentration.

In the case of the electrochemical impedance spectroscopy, the inhibition efficiency is calculated using charge transfer resistance as follow²⁹:

$$P \% = \frac{(R_{ct(inh)} - R_{ct})}{R_{ct(inh)}} \times 100 \quad (4)$$

Where, R_{ct} and $R_{ct(inh)}$ are the charge transfer resistance values in the absence and presence of inhibitor for C steel in 1M HCl, respectively.

As the inhibitor concentration increased, the R_{ct} values increased, but the C_{dl} values tended to decrease.

The decrease in C_{dl} value is due to the adsorption of inhibitor on the metal surface³⁰.

The inhibition efficiency increases with increasing inhibitor concentration. This fact suggests that the inhibitor molecules may first be adsorbed on the steel surface and cover some sites of the electrode surface. These layers protect steel surface.

Weight Loss Measurements

Effect of Inhibitor Concentrations

The corrosion parameters such as weight loss (ΔW), rate of corrosion (k), surface coverage (θ) and inhibition efficiency (p) for Domperidone inhibitor are listed in Table 3. The weight losses given by equation:

$$\Delta W = (W_1 - W_2) \quad (5)$$

Where, W_1 and W_2 are the weight of specimen before and after the reaction, respectively.

In all cases the increase of inhibitor concentrations was accompanied by a decrease in weight loss and an increase in the percentage inhibition³¹. These results lead to conclusion that, the compound under investigation is fairly efficient as inhibitor for C-steel dissolution in hydrochloric acid solution. To clear up the influence of this inhibitor on the mechanism of inhibition, the corrosion rate (k) was calculated using the following equation³²:

$$k = \frac{(W_{\text{free}} - W_{\text{inh}})}{A} \quad (6)$$

Where, k is the corrosion rate, W_{inh} and W_{free} are the weights loss of specimen in presence and absence of inhibitor respectively, A is the surface area in cm^2 and t is the time in hours.

The degree of surface coverage (θ) by the adsorbed molecules was calculated from equation²⁸:

$$\theta = \frac{(W_{\text{free}} - W_{\text{inh}})}{W_{\text{free}}} \quad (7)$$

It was found that the degree of surface coverage (θ) of the inhibitor increases by increasing the inhibitor concentration.

The inhibition efficiencies ($P\%$) of drug were determined from equation³⁴:

$$P\% = (\theta) \times 100 \quad (8)$$

Effect of temperature

The effect of rising temperature on both corrosion and corrosion inhibition of C-steel in 1M HCl solution at different temperatures was studied in range (25-55 °C) and this investigated by weight loss measurements.

The corrosion parameters such as weight loss (ΔW), rate of corrosion (k), surface coverage (θ) and inhibition efficiency ($P\%$) for 500 ppm from Domperidone listed in Table 4.

Kinetic Parameters

Activation Energy

The apparent activation energy, E_a , for the corrosion of C-steel sample in 1M HCl solution in the absence and presence of different concentrations of drug, at 25, 35, 45, and 55 °C were calculated from Arrhenius equation.

$$k = A e^{(-E_a/RT)} \quad (9)$$

The logarithmic form:

$$\ln k = \ln A - E_a / RT \quad (10)$$

Where, k is the corrosion rate, A is the Arrhenius constant, R is the gas constant and T is the absolute temperature.

Arrhenius plots of $\ln k$ vs $1/T$ in the absence and presence of different concentrations of the used inhibitor where shown graphically in Figure 3 give straight lines with slope of $(-E_a/R)$. Activation energies were calculated and give rise the values of E_a increase in case of inhibitor concentrations from 100-500 ppm (56.59 - 79.26) compared with blank (47.38). The data shows that the reaction of inhibitors is a typical physical adsorption for CPBP onto C-steel.

The change in enthalpy and entropy of activation values (ΔH^* , ΔS^*) were calculated from the transition state theory³⁵.

$$k = RT/Nh \exp(\Delta S^*/R) \exp(-\Delta H^*/RT) \quad (11)$$

Where, h is the Plank constant, N is the Avogadro's number, and R is the ideal gas constant.

A plot of $\log(k/T)$ versus $1/T$, gave straight lines for carbon steel dissolution in 1M HCl in the absence and presence of different concentrations of inhibitors. Straight lines are obtained with a slope of $\Delta H^*/R$ and an intercept of $\log(R/Nh) + \Delta S^*/R$. The values of ΔH^* and ΔS^* were calculated from the above equation and show that the values of ΔH^* (KJmol^{-1}) increase from 19.44 (blank) into 34.16 (CPBP) in the positive direction indicate that the reaction is endothermic reaction. On the other hand, the values of ΔS^* ($\text{KJmol}^{-1}\text{K}^{-1}$) show the negative charges direction indicate that decrease in disordered.

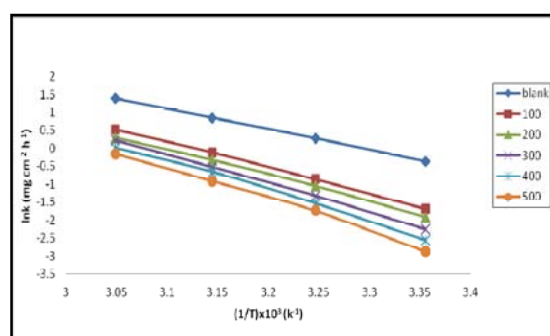


Figure 3: Arrhenius plots ($\ln k$ vs. $1/T$ curves) for carbon steel dissolution in absence and presence of different concentrations of CPBP in 1M HCl solution.

Adsorption Isotherm

The type of adsorption isotherm and the effect of temperature on the corrosion rate were studied. It is generally accepted that the studied drug, compound inhibit the corrosion process by adsorbing at the metal/solution interface³¹.

In addition, it is believed that the formation of a solid organic molecule complex with the metal atom has received considerable attention³².

The equation that fits our results is that due to Langmuir isotherm and it given by the general equation:

$$C/\theta = 1/K_{ads} + C \quad (12)$$

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

The degrees of surface coverage (θ) for different concentrations of inhibitor in 1M HCl solution have been calculated from weight loss measurements by using equation (7).

In these cases, the plots of C/ θ versus C yield a straight line with intercept of (1/ K_{ads}) and with slope approximately equal unity was obtained, Figure 4.

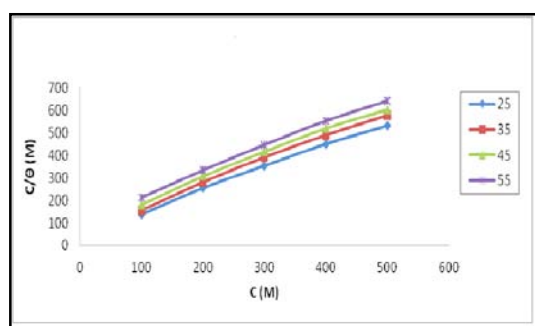


Figure 4: Langmuir isotherm adsorption model on the carbon steel surface of CPBP in 1M HCl at different temperatures.

The small deviation from unity is generally attributed to the interaction of the adsorbed inhibitor molecules on heterogeneous carbon steel surface. This indicates that, the adsorption of inhibitor on the carbon steel surface in 1M HCl solution follows Langmuir's adsorption isotherm. The free energy of adsorption (ΔG_{ads}) at different temperatures was calculated from the following equation³¹:

$$-\Delta G_{ads} = RT \ln (55.5 K_{ads}) \quad (13)$$

Where, C is the inhibitor concentration, θ is the fraction of the surface covered, K_{ads} is the equilibrium constant of the inhibitor adsorption process, the value 55.5 is the molar concentration of water in solution in mol dm⁻³, R is the gas constant, T is the absolute temperature and ΔG_{ads} is the standard free energy of adsorption.

The heat of adsorption (Q_{ads}), which is obtained from the slopes of the straight lines when K_{ads} plot versus 1/T, is equal to $-Q_{ads}/R$. Since the pressure is a constant, Q_{ads} is equal to enthalpy of adsorption (ΔH_{ads}) with good approximation. Entropy of inhibitor adsorption (ΔS_{ads}) can be calculated using the following equation:

$$\Delta G_{ads} = \Delta H_{ads} - T \Delta S_{ads} \quad (14)$$

The calculated values of the adsorption free energy, ΔG_{ads} , adsorption entropies, ΔS_{ads} , and adsorption enthalpies, ΔH_{ads} , are given in Table 5. The values of ΔG_{ads} indicated that the adsorption mechanism of the inhibitor

on carbon steel in 1 M HCl solution is a mixed type adsorption.

Table 5: Thermodynamic parameters of adsorption on carbon steel surface in 1M HCl containing different concentrations of CPBP at different temperatures

Temp., °C	$K_{ads} \times 10^3 M^{-1}$	$\Delta G_{ads} KJ mol^{-1}$	$\Delta H_{ads} J mol^{-1}$	$\Delta S_{ads} JK mol^{-1}$
25	2.2	-33.8	68.74	114.79
35	1.56	-33.45		109.92
45	1.16	-33.78		107.5
55	1.12	-34.178		105.44

CONCLUSION

Based on the obtained results, the following conclusions are accomplished:-

The used expired drug is excellent inhibitor and act as the mixed type inhibitors for carbon steel corrosion in hydrochloric acid solution.

Inhibition efficiencies increased by increasing inhibitor concentration and temperatures up to 328 K.

The uniform increasing inhibition efficiency as the function of concentration and the adsorption of the additive obeys the Langmuir adsorption isotherm.

All entropy parameters for adsorption of inhibitor molecules on carbon steel are positive and increased by increasing the temperature which indicates that the inhibitor being more oriented and less ordered on the surface of the metal.

Thermodynamic parameters of the adsorption (E^* , ΔH^* and ΔS^*) were calculated and showed that the used inhibitor decreased the rate of corrosion.

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