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Inhibitory action of Thiazole and Triazole on mild Steel Corrosion in Hydrochloric acid Solutions

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Abstract

Thiazole [3-(piperazin-1-yl)benzo(d)isothiazole] and Triazoles (Hydroxybenzotriazole) were used as corrosion inhibitors for mild steel in hydrochloric acid solution. The inhibition efficiency depended on the concentration and type of the thiazole and triazoles. The inhibition efficiency ranged between 66 to 77 % at the highest concentration (25 mM), and between 33 to 44% at the lowest concentration (5 mM) of inhibitor in 1 M HCl solution. Inhibition efficiency decreased with rise in temperature, this corresponded to surface coverage of the metal by the inhibitor. The results also showed that, the inhibitors were adsorbed on the mild steel surface according to Langmuir, Temkin's and Frundlich adsorption isotherm.

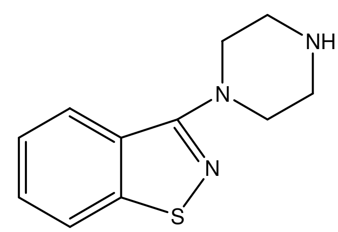
Keywords: Corrosion, Mild steel, Hydrochloric acid, thiazole, triazole, Langmuir adsorption, Temkin's adsorption isotherm.

Introduction

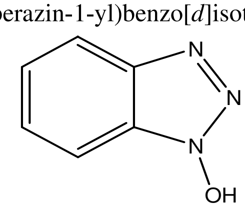
One of the most serious problems in the world, especially in an industry is corrosion which can be defined as the degradation of a material's properties or mass over time due to environmental effects. For most metallic materials, the natural tendency of material's compositional elements to return to their most thermodynamically stable state of formation of oxides or sulfides, or other basic metallic compounds generally considered to be ores with slow process rate. Under normal circumstances, iron and steel corrode in the presence of both oxygen and water. The rate of corrosion is increased by the acidity or the velocity of the water, by the motion of the metal, by an increase in the temperature or aeration, by the presence of certain bacteria, or by other less prevalent factors[1].

Using inhibitors is an effective method for corrosion to control the corrosion of metals. Inhibitors are compounds that control corrosion processes of metals. Many studies on inhibitors have been carried out among them nitrogen (N) containing inhibitor is one of the focuses of the studies[2-18]. Many researchers have investigated many N containing inhibitors for the corrosion inhibition of metal in acid solutions, N containing organic inhibitor acts as a strong inhibitor for metal in acid[19-28]. Thiazole and triazole-type compounds containing N and S heteroatom. Heterocyclic compound on the corrosion inhibition of metal in acidic media have attracted more attention because of their excellent corrosion inhibition performance[29-32]. The researches by Desai and Indorwala showed that some Triazoles used as a Corrosion inhibitor for mild steel in Hydrochloric Acid and the inhibition effect was found very excellent.

However, Wang et al. also investigated the effect of some mercaptotriazole derivatives synthesized containing different hetero atoms and substituents in the organic structures on the corrosion and hydrogen permeation of mild steel in HCl solution and their results revealed that all the mercaptotriazole derivatives performed excellently as corrosion inhibitors. Especially, some N-and S- containing triazole derivatives are environmentally friendly corrosion inhibitors compared with some commercial acid corrosion inhibitors, which are highly toxic, such as chromate and nitrite[33-34]. Therefore, In this work, the inhibition ability of PBIT [3-(piperazin-1-yl) benzo (d) isothiazole] and HOBT (hydroxyl benzotriazole) for mild steel corrosion in HCl solution at various temperatures was evaluated by the gravimetric method. The inhibitory mechanism of these organic compounds was discussed based on the analysis of the adsorption isotherm.



3-(piperazin-1-yl)benzo[*d*]isothiazole



Hydroxy benzotriazole

Material and Methods

Mild steel contains Fe = 99.746; Mn = 0.100; C = 0.058; Al =

0.033; Si = 0.010; Cr = 0.008; Cu = 0.004; Ni = 0.0029; Mo = 0.002% were used in this study. Each sheet, which was 0.12 cm in thickness, was mechanically press-cut into coupons of dimension 3.5 x 3 cm with small hole of about 5 mm diameter near the upper edge. These coupons were used in the "as cut" condition, inhibition efficiency without further polishing, but were de-greased in absolute ethanol, dried in acetone, weighed and stored in a moisture-free desiccator prior to use. All chemicals and reagents used were of analytical grade and used as source without further purification. The aggressive media were, respectively, 1, 2 and 3M HCl solution. Triazoles were used as inhibitors in the concentration range 5, 10, 15, 20 and 25 mM.

The test specimens were immersed in 1.0, 2.0 and 3.0 M HCl solution with without inhibitors. Only one specimen was suspended by a glass hook, in each beaker containing 230 ml of the test solution and was open to air at room temperature for 24 h. After the immersion period, the surface of specimen was cleaned by double distilled water followed by rinsing with acetone and sample was weighed again in order to calculate inhibition efficiency (η %). Triplicate experiments were performed in each case and the calculated mean values of the weight loss data and inhibition efficiency and which was shown in figure-1 (a and b).

To study the effect of temperature on corrosion rate, the specimen were immersed in 230 ml of 1.0 M HCl solution with various triazoles as inhibitor having concentration 5, 10, 15, 20 and 25 mM at solution temperatures of 313, 323 and 333 K for a period of 3h. To study the effect of temperature, thermostat assembly with accuracy of + 0.5^{o}C was used. From the weight loss data calculated the inhibition efficiency (I.E. %), energy of activation (E_a), heat of adsorption (Q_{ads}), free energy of adsorption (ΔG^o_{ads}), enthalpy of adsorption (ΔH^o_{ads}) and entropy of adsorption (ΔS^o_{ads}).

Results and Discussion

The results are presented in table-1 and figures-1 to 6. To assess the effect of corrosion of mild steel in HCl, PBIT and HOBT were used as inhibitors. The corrosion rate of mild steel in HCl was increased with the acid concentration which was shown in figure-1, 2. The inhibition efficiency (η %) and degree of surface coverage (θ) at each concentration of PBIT and HOBT was calculated by comparing the corrosion loss in the absence (W_u) and presence of inhibitors (W_i) using the relationships:

$$\eta\% = \left(\frac{Wu - Wi}{Wi}\right) X100 \quad (1)$$

$$\theta = \left(\frac{Wu - Wi}{Wi}\right) \quad (2)$$

As a constant inhibitor concentration, the inhibition efficiency decreased with the increase in acid concentration. At 25 mM inhibitor concentration, the inhibition efficiency of PBIT [3-(piperazin-1-yl)benzo(d)isothiazole] was 66.01, 52.15 and 41.59 with respect to 1.0, 2.0 and 3.0 M acid concentrations respectively. At a constant acid concentration, the inhibition efficiency of the triazole increased with the inhibitor concentration, e.g. with HOBT in 1.0 M HCl the inhibition efficiency was found to be 43.79, 51.63, 59.48, 66.01 and 76.47 with respect to 5, 10, 15, 20 and 25 mM inhibitor concentrations respectively (figure 1-a, 1-b,).

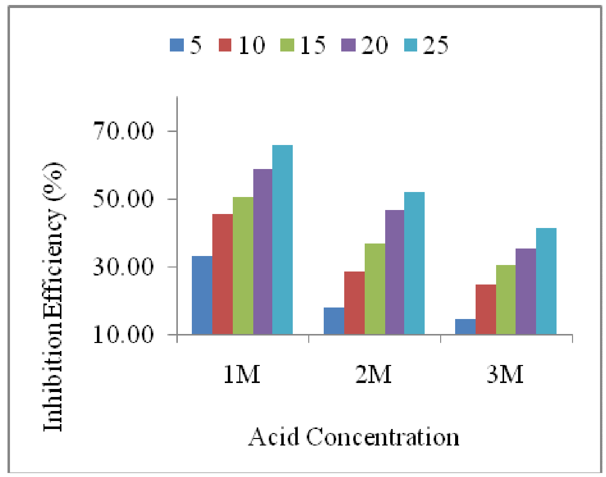


Figure-1(a)
Effect of inhibition efficiency of Thiazole for mild steel at different acid and inhibitor concentrations for 24 h at 301 K

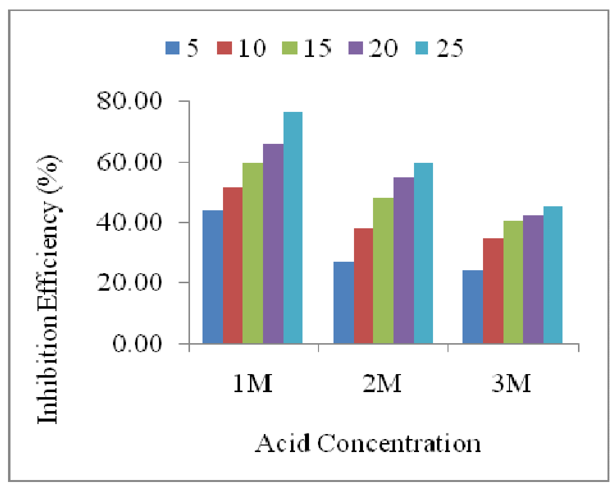


Figure-1(b)
Effect of inhibition efficiency of Triazoles for mild steel at different acid and inhibitor concentrations for 24 h at 301 K

The temperature had significant influence on the metal corrosion rates. The effect of change in temperature on the corrosion rates of mild steel in 1.0 M HCl was examined. It was found that, the corrosion of mild steel increased with the increase in temperatures (table-2). Corrosion rate was measured in 1.0 M HCl containing 5, 10, 15, 20 and 25mM inhibitor concentration at a solution temperature of 313, 323 and 333 K for an immersion period of 3 h. In 1.0 M HCl solution with 25 mM Hydroxy benzotriazole concentration, the inhibition efficiency was found to be 76.92, 75.00 and 74.53 at 313, 323 and 333 K respectively (figure-2a, 2b).

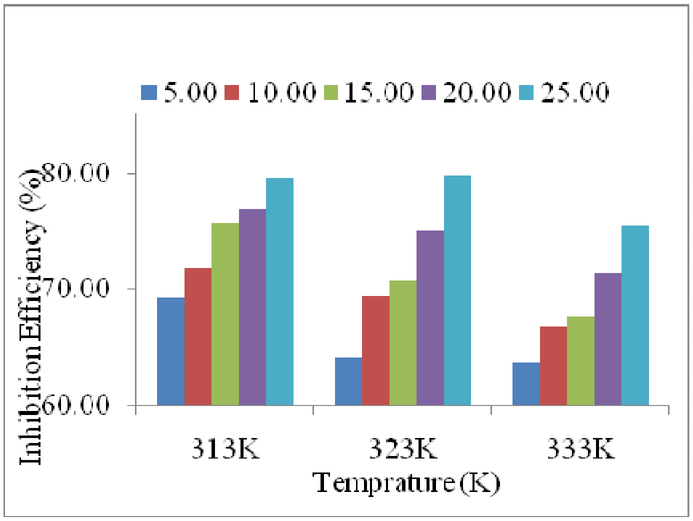


Figure-2 (a)
Effect of inhibition concentration of PBIT for mild steel in 1M HCl at different temperatures for 3h

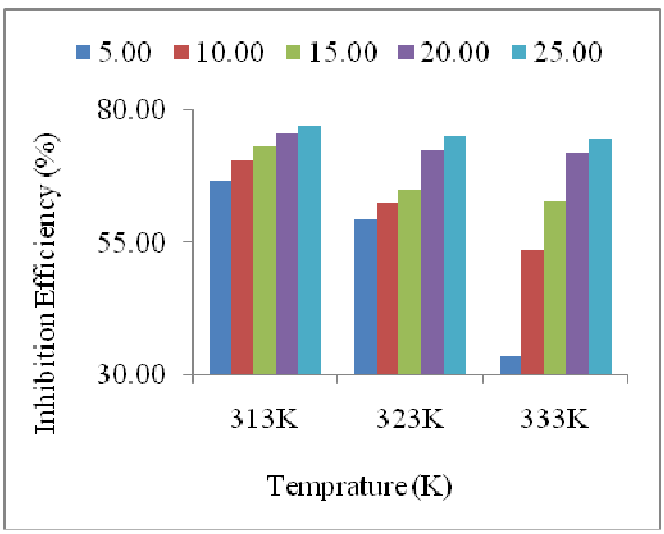


Figure -2 (b)
Effect of inhibition concentration of HOBT for mild steel in 1M HC lat different temperatures for 3h

In the present study general type of corrosion occurs predominately and less pitting. Plotting of log θ/1-θ versus log C (%), straight lines were obtained, indicating that the adsorption of the added inhibitors followed the Langmuir adsorption isotherm (figure-3a, 3b). Therefore, adsorption of these compounds is assumed to occur uniformly over the metal surface.

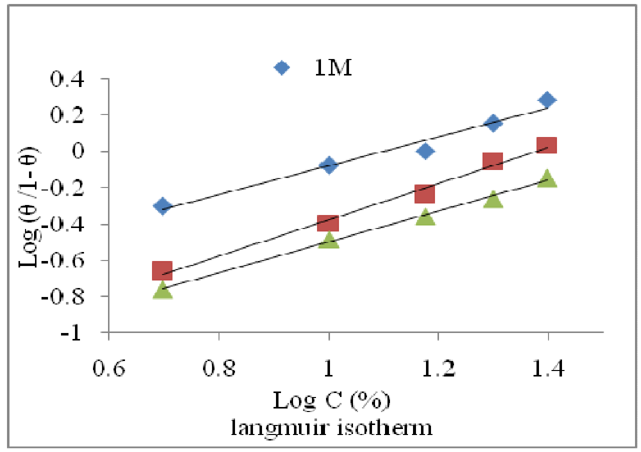


Figure-3(a)
Plot of log (θ /1- θ) versus log C for PBIT at different concentrations of HCl for 24 h at 301 K

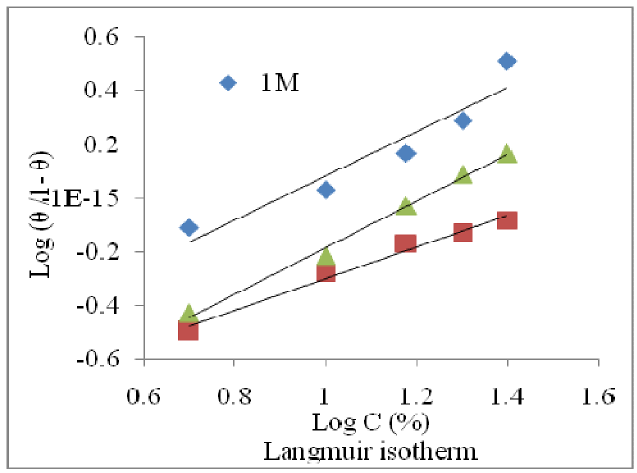


Figure-3(b)
Plot of log (θ /1- θ) versus log C for HOBT at different concentrations of HCl for 24 h at 301 K

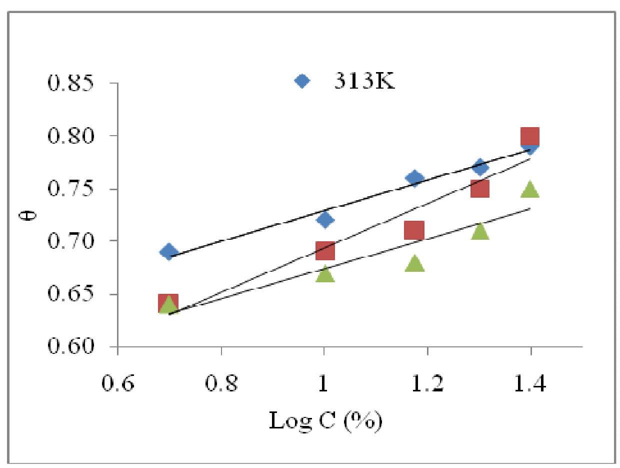


Figure-4(a)
Plot of θ versus log C for PBIT at different concentrations of HCl for 3h

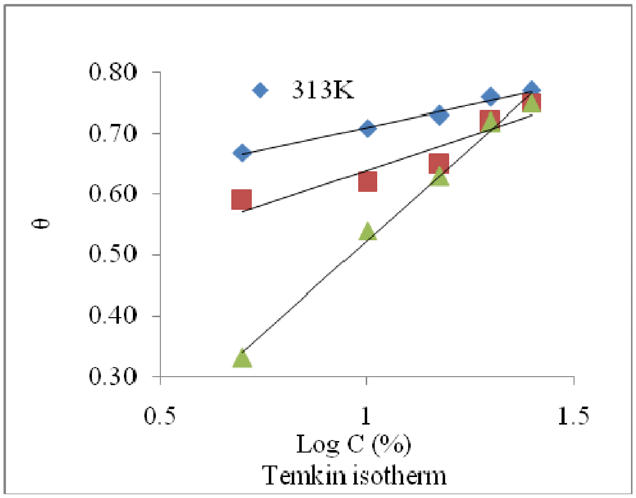


Figure-4(b)
Plot of θ versus log C for HOBT at different concentrations of HCl for 3h

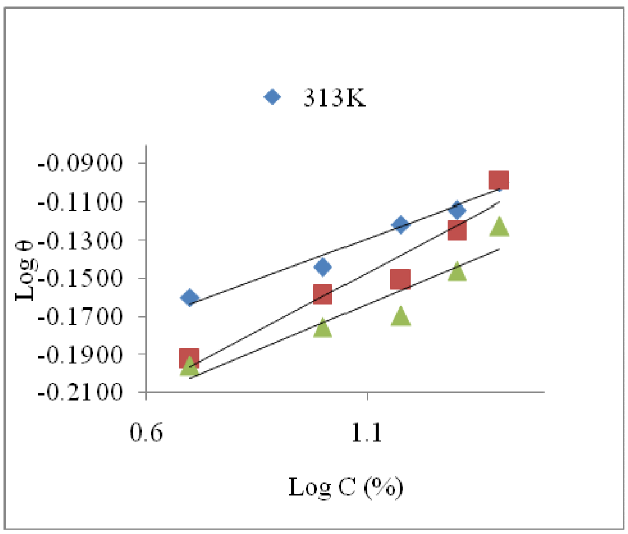


Figure-5(a)
Plot of log θ versus log C for PBIT at different concentrations of HCl for 3h

Plot a graph θ versus log *C* gives straight line (figure-4 a and b) showing that the adsorption of the compound on the mild steel surface from 1 M HCl obeys also Temkin's adsorption isotherm and also obey Freundlich adsorption isotherm.(figure-5 a and b) It is also found that the degree of adsorption of the inhibitors increases with their concentration.

The values of the free energy of adsorption (ΔG^0_{ads}) were calculated with the slope of the following equation[35].

$$LogC = Log\left(\frac{\theta}{1-\theta}\right) - LogB \qquad (3)$$

Where: $LogB = -1.74 - \left(\frac{\Delta G^0_{ads}}{2.303RT}\right)$ and C is the inhibitor concentration. The mean ΔG^0_a values are negative almost in all cases and lie in the range of -5.14 to -6.77 kJ. mol^{-1} shown in table-1. The most efficient inhibitor shows more negative ΔG^0_a value. This suggests that they be strongly adsorbed on the metal surface[36]. The values of heat of adsorption (Q_{ads}) were calculated by the following equation.

$$Q_{ads} = 2.303R\left[Log\left(\frac{\theta_2}{1-\theta_2}\right) - Log\left(\frac{\theta_1}{1-\theta_1}\right)\right] X \left[\left(\frac{T_1T_2}{T_2-T_1}\right)\right] \qquad (4)$$

From table -1, it is evident that in all cases, the (Q_{ads}) values are negative and ranging from 1.32 to -96.65 kJ. mol^{-1}. The negative values show that the adsorption, and hence the inhibition efficiency, decreases with a rise in temperature[37].

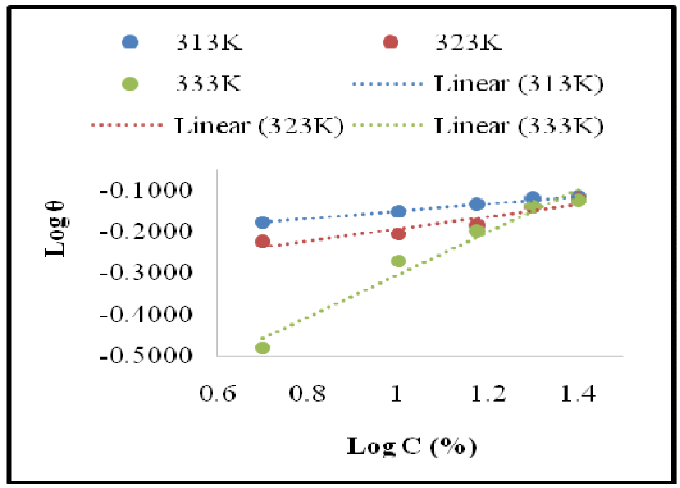


Figure-5(b)
Plot of log θ versus log C for HOBT at different concentrations of HCl for 3h

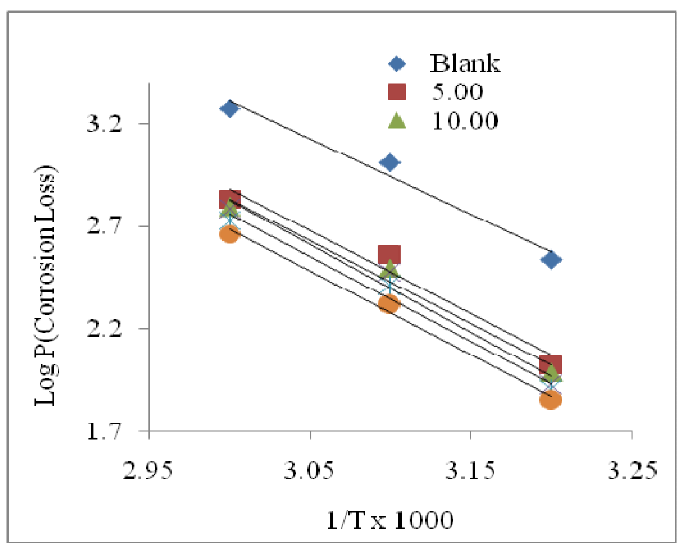


Figure-6(a)
Arrhenius plots for corrosion of mild steel in 1 M HCl in absence and presence of Triazoles

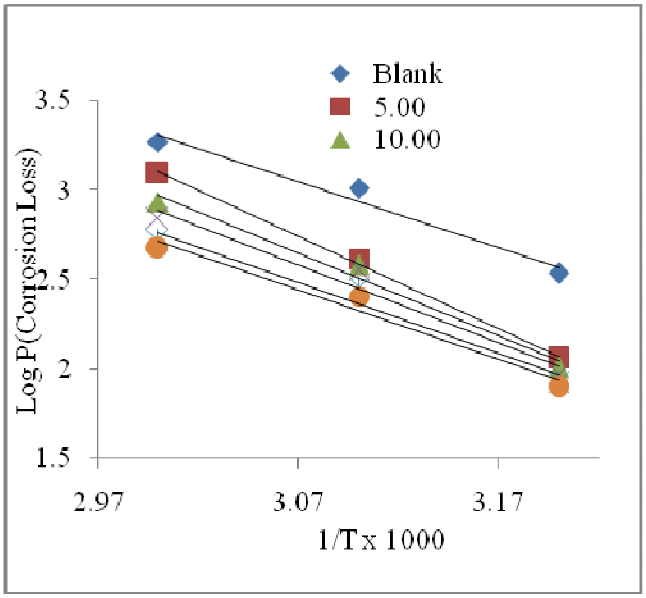


Figure-6(b)
Arrhenius plots for corrosion of mild steel in 1 M HCl in absence and presence of Triazoles

Mean 'E_a' value was calculated by using equation (5) for mild steel in 1M HCl is 72.79 kJ.mol^{-1} while in acid containing inhibitor, the mean E_a values are found to be higher than that of an uninhibited system (table-1). Higher values of $E_a$in the presence of the triazoles which acts as inhibitor is a good indication of strong inhibiting action of the triazoles by increasing the energy barrier for the corrosion process. Higher values of E_a in the presence of extract can also be correlated with the increase in thickness of the double layer that enhances the E_a of the corrosion process[38]. The values of $E_a$calculated from the slope of an Arrhenius plot (figure-6a,6b) and using equation (5) are almost similar. Energy of activation (E_a) has been calculated from the slopes of log p versus 1/T (p = corrosion rate, T = absolute temperature) and also with the help of Arrhenius equation.

$$Log\frac{P_2}{P_1} = \frac{Ea}{2.303R}\left[\left(\frac{1}{T_1}\right)-\left(\frac{1}{T_2}\right)\right] \quad (5)$$

Where p_1 and p_2 are the corrosion rate at temperature T_1 and T_2 respectively.

The enthalpy of adsorption (ΔH^0_{ads}) and entropy of adsorption (ΔS^0_{ads}) were calculated using the following equation-6 and 7.

$$\Delta H^0 ads = Ea - RT \quad (6)$$

$$\Delta S^0{}_{ads} = \frac{\Delta H^0{}_{ads} - \Delta G^0{}_{ads}}{T} \quad (7)$$

The enthalpy changes (ΔH^0_a) are positive, indicating the endothermic nature of the reaction suggesting that higher temperature favors the corrosion process[39]. The entropy (ΔS^0_a) values are positive, confirming that the corrosion process is entropically favorable[40].

The inhibitory mechanism is a separation process involving i. the inhibitor is adsorbed on the surface of the metal forming a compact protective thin layer and ii. the inhibitor forms a precipitate on the surface of the metal, acting on the aggressive media to form protective precipitates or remove the aggressive agents[41]. Adsorption, on the other important notes can be described by two main types of interaction, which are physisorption and chemisorptions[42].

Most 0rganic inhibitors are compounds with at least one polar unit, having atoms of nitrogen, oxygen, sulfur and in some cases, selenium and phosphorus. Amine type inhibitors have electron-donating ability and their action is attributed to the adsorption of the inhibitor molecules on the metal surface through an unshared pair of electrons belonging to the nitrogen atom[43]. The same is applicable to sulphur and oxygen atoms. It seems reasonable to suppose that the presence of two polar groups ensure better attachment than single one, but this view is not universally observed. In any case, it might be asked whether two nitrogen groups would not be as sufficient as one nitrogen and one sulfur group; perhaps the answer that the two different elements serve to provide attachment at different sites, that they attach themselves to the surface probably by the nitrogen or sulfur atom, is generally accepted.

Table-1
Energy of activation (E_a), heat of adsorption (Q_{ads}) and free energy of adsorption (ΔG^0_{ads}) for mild steel in 1.0 M HCl Containing inhibitors

System	Mean Ea from Eq.(2) (kJ. mol^{-1})	Ea from Arrhenius plot (kJ. mol^{-1})	Q_{ads} (kJ. mol^{-1})		Mean value (kJ. mol^{-1})		
			313-323 (K)	323-333 (K)	ΔG^o_{ads}	ΔH^o_{ads}	ΔS^o_{ads}
Blank	72.79	70.38	-	-	-	-	-
3-(piperazin-1-yl) benzo(d)isothiazole	80.81	77.81	1.32	-22.03	-5.56	78.17	0.2643
Hydroxy benzotriazole	76.99	74.52	-8.86	-2.24	-5.14	74.35	0.2510

Conclusion

As a constant inhibitor concentration, the inhibition efficiency of all inhibitors decreases as the concentration of acid increases. At all concentration of acid, as the inhibitor concentration increases inhibition efficiency increases and corrosion rate decreases. As the temperature increases corrosion rate increases in plain acid. Addition of inhibitors in corrosive media indicates that as the temperature increases corrosion rate increases while inhibition efficiency decreases.

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