Comparative Study of Corrosion Inhibition Capacities of Seroquel and Clotrimazole Drug Intermediates on Zinc Metal Surface in Acidic Corrosive Medium.

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Abstract— Corrosion inhibition capacities of two drug intermediates, seroquel and clotrimazole on zinc surface were compared. The comparison was done using the data obtained by experimental methods like mass loss, EIS and Tafel polarization. The metal surface was examined by SEM studies. The results obtained by the three experimental methods were in good agreement in case of both the drug intermediates. The seroquel was found to follow Temkin adsorption isotherm and its corrosion inhibition property was found to be very good in case of higher temperature. The clotrimazole followed Langmuirs' adsorption isotherm and it showed an excellent corrosion inhibition at ambient temperature. Both the drug intermediates mitigate corrosion by mixed type of inhibition mechanism involving both physical and chemical adsorption on the zinc metal surface. Further SEM micrographs confirmed the effectiveness of both the drug intermediates in controlling the rate of zinc corrosion.

Index Terms—corrosion inhibitor, clotrimazole, drug intermediate, EIS, seroquel, SEM, Tafel polarization.

1 INTRODUCTION

Corrosion is a generally a slow but strong spontaneous deterioration of materials due to the action of surrounding corrosive environment by chemical or electrochemical means. In this work we have discussed about corrosion of metals or alloys though non metals also undergo split, crack or decay. Metals keep loosing its properties because of corrosive reactions with its environment. Metals usually available in nature in stable combined state like oxide, sulfide, carbonate, etc. Because of this they have low energy and hence thermodynamically stable. Considerable amount of energy is invested in extracting metals so these metals have high energy and thermodynamically unstable. Due to this reason extracted metals always try to get back to its thermodynamically stable state through these corrosion processes by loosing properties like strength, ductility, etc. These corrosion reactions effects industries by increasing cost of maintenance, decrease the rate of production and efficiency, replacement of corroded articles, etc. So the control of this process is of high value.

There are different methods available to tackle the corrosion, but the use of corrosion inhibitors had remained one of the most economical and effective method [1, 2]. In this method a small quantity of chemicals (inhibitor) is added to corrosive environment, which intern interact with the metal/alloy surface by physically and/or chemically and reduces the rate of corrosion. Based on the mechanism of inhibition the inhibitors are classified as anodic, cathodic and mixed inhibitors. The efficiency of inhibition is increased by the presence of certain halogens and also hetero atoms like nitrogen, sulfur, oxygen, phosphorous [3, 7]. Major inhibitors used in industries are toxic in nature because of this use of drug intermediate as a potent corrosion inhibition is an exciting prospect. Zinc is an active metal which is extensively used in industries. In the present work we have compared corrosion inhibition efficiencies of the two drug intermediate namely seroquel and clotrimazole on the zinc surface. The corrosion inhibition capacities of these drug intermediates were compared on the basis of experimental methods such as mass loss, potentiodynamic polarization, and impedance spectroscopic studies.

2 EXPERIMENTAL

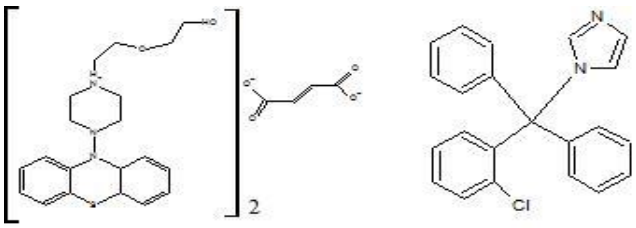
2.1 Material and corrosive medium preparation

Zinc metal sheets were manually cut into desired size and shape for the experimental studies. All the zinc samples were immersed in dilute acid solution and then abraded with SiC emery papers of grades from 180 to 2000 to get a mirror finish. Afterwards the samples were rinsed with acetone then thoroughly cleaned with deionized water, dried and stored in a dessicator. The corrosive acid medium used for the comparative study was 0.1 M hydrochloric acid, which was prepared by diluting concentrated AR grade hydrochloric acid with deionized water.

2.2 Inhibitor

Fig.1 Molecular structure of a) seroquel b) clotrimazole.

a



The different concentrations of seroquel drug intermediate solution was prepared by weighing desired weight of the drug and dissolving and diluting it by using 0.1 M HCl solution. The different concentrations of clotrimazole drug intermediate solution was prepared by weighing desired weight of the drug, dissolving it in absolute ethanol and diluting it using 0.1 M HCl solution. Molecular structure of seroquel and clotrimazole is shown in Fig. 1a and b respectively.

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2.3 Mass loss study

The surface cleaned zinc samples having a dimension of 4 X 2 X 0.1 cm^2 used for mass loss study were weighed before the experiment. In this method the zinc metal samples were suspended inside the 100 ml 0.1 M HCl medium without and with differential inhibitor concentrations. The samples removed after stipulated time duration were washed with deionized water, rinsed with acetone, dried and weighed. The corrosion rate (v_{corr}), inhibition efficiencies ($\%\eta_m$) and surface coverage (θ) was calculated by Eqs. (1), (2) and (3) respectively.

$$v_{corr} = \frac{\Delta m}{ST} \quad -----(1)$$

Where, Δm is the mass loss difference without and with drug intermediates, S is the surface area and T is the time of exposure in hours.

$$\%\eta_m = \frac{v^o_{corr} - v_{corr}}{v^o_{corr}} \times 100 \quad -----(2)$$

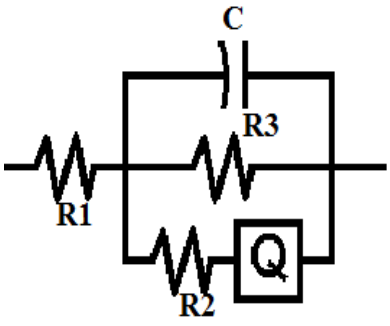
Where v^o_{corr} and v_{corr} are the corrosion rates of zinc in the absence and presence of seroquel, respectively.

$$\theta = \frac{\%\eta_m}{100} \quad -----(3)$$

2.4 Polarization and impedance studies

The surface cleaned zinc samples having an exposed area of 1cm2 were used in electrochemical studies for both the drug intermediates. Electrochemical workstation CHI 608D (CH Instruments, Austin, USA) having three electrode system with zinc as working, calomel as reference and platinum as auxiliary electrode were used for the present study. The three electrodes were immersed in solution without and with different drug intermediate concentrations and connected to electrochemical work station. This assembly was made to stand for 10 minutes before each estimation to attain stable ocp.

2.4.1 Electrochemical impedance studies



R1, R2, R3 - Resistance,
C - Capacitance ,
Q - Constant phase element.

Fig. 2 Electrical equivalent circuit used for impedance data fitting for both the drug intermediates.

EIS study was done at a frequency range from 10 kHz to 1 kHz at 0.005 Vs^{-1}. The percentage inhibition efficiency ($\%\eta_z$) and double layer capacitance values (C_{dl}) were calculated using the Eqs. 4 and 5.

$$\%\eta_z = \frac{R_p - R^o_P}{R_P} \times 100 \quad -----(4)$$

Where, R_p and R^o_p are the polarization resistance values with and without inhibitor respectively.

$$C_{dl} = \left(QR_{ct}^{1-n}\right)^{\frac{1}{n}} \quad -----(5)$$

Where Q is the constant phase element (CPE) ($\Omega^{-1} S^n cm^{-2}$) and n is the CPE exponent.
The experimental curves were fitted to equivalent electrical circuit shown in Fig. 2 using ZSimpWin 3.21 and parameters obtained were tabulated.

2.4.2 Tafel polarization studies

Polarization measurements were recorded at a scan rate of 0.01 mVs^{-1} in the potential range ± 0.2 V at open circuit potential (ocp). The percentage inhibition efficiency $\%\eta_p$ was calculated from the Eq. 6.

$$\%\eta_p = \frac{i^o_{corr} - i_{corr}}{i^o_{corr}} \times 100 \quad -----(6)$$

Where, i_{corr} and i^o_{corr} are the corrosion current values with and without inhibitor respectively.

2.5 Scanning electron microscope studies

This experimental study was done to know more about surface appearance of zinc metal before and after exposing it into a known concentration of drug intermediates. For this study surface cleaned metal samples were suspended inside a beaker containing 100 ml 0.1 M HCl solution without and with a 1000 ppm seroquel and 500 ppm clotrimazole drug intermediates for a stipulated time duration. After removal, zinc samples were cleaned with deionized water, acetone, then dried and kept inside a dessicator till their usage for analysis. The SEM images for seroquel and clotrimazole drug intermediates were taken using VEGA3 TESCAN and ZEISS EVO MA18 respectively.

3 Results and Discussion

3.1 Mass loss studies

The experimental parameters obtained by the mass loss study were presented in Table 1. The variation of inhibition efficiencies with respect to change in inhibitor concentration is shown in Fig. 3.
The results obtained showed a decrease in mass loss of the zinc metal samples in the presence of both the drug intermediates when compared to their absence in corrosive medium. Corrosion rate of zinc sample in only 0.1 M HCl solution was found to be more than the sample in 0.1 M HCl with drug intermediates. Accordingly the corrosion inhibition efficiency was high in presence of drug intermediates with increased concentrations.

	C_{inh} (ppm)	(ρ) (g/cm² h)	η_m (%)	θ
Seroquel	Blank	0.0106	-	-
	200	0.0071	33.01	0.33
	400	0.0062	41.50	0.41
	600	0.0061	42.45	0.42
	800	0.0049	53.77	0.53
	1000	0.0042	60.37	0.60

	C_{inh} (ppm)	(ρ) (g/cm² h)	η_m (%)	θ
Clotrimazole	Blank	2.653	-	-
	100	1.328	49.94	0.49
	200	0.965	63.60	0.63
	300	0.828	68.78	0.68
	400	0.665	74.91	0.74
	500	0.584	77.97	0.77

Table 1 Mass loss studies of zinc metal in 0.1 M HCl solution without and with seroquel and clotrimazole drug intermediates.

From Table 1 it is clear that both the drug intermediates retard the rate of corrosion in their presence in 0.1 M HCl solution. But the clotrimazole showed a higher corrosion inhibition efficiencies compared to the other drug intermediate at ambient temperature. Hence it indicates, clotrimazole is a better corrosion inhibitor for zinc in 0.1 M HCl solution at ambient temperature.

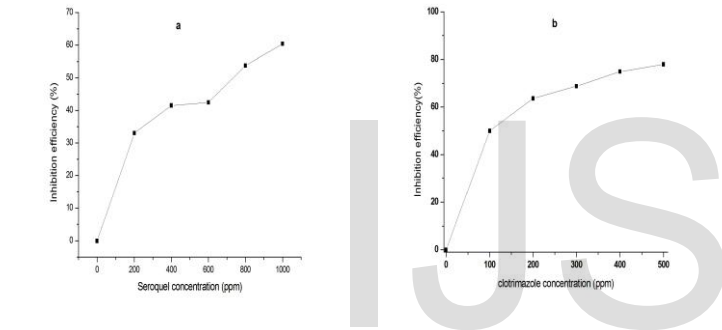


Fig. 3 Variation of inhibition efficiency with inhibitor concentration.

Inhi.	C_{inh} (ppm)	R_p Ω cm²	C_{dl} F/cm²	Goodness of fit (x 10^{-4})	%η_z	θ
Seroquel	Blank	279	0.0021	2.641	-	-
	200	439	0.0066	2.998	36.44	0.36
	800	1050	0.0140	1.416	73.39	0.73
	1000	1266	0.0145	0.852	77.93	0.78
Clotrimazole	Blank	305	0.0128	0.4406	-	-
	100	811	0.0081	0.3044	62.40	0.62
	300	2151	0.0025	2.5010	85.82	0.85
	500	3131	0.0029	3.1860	90.25	0.90

3.2 EIS studies

Table 2 EIS studies of zinc metal in 0.1 M HCl solution without and with seroquel and clotrimazole drug intermediates at room temperature.

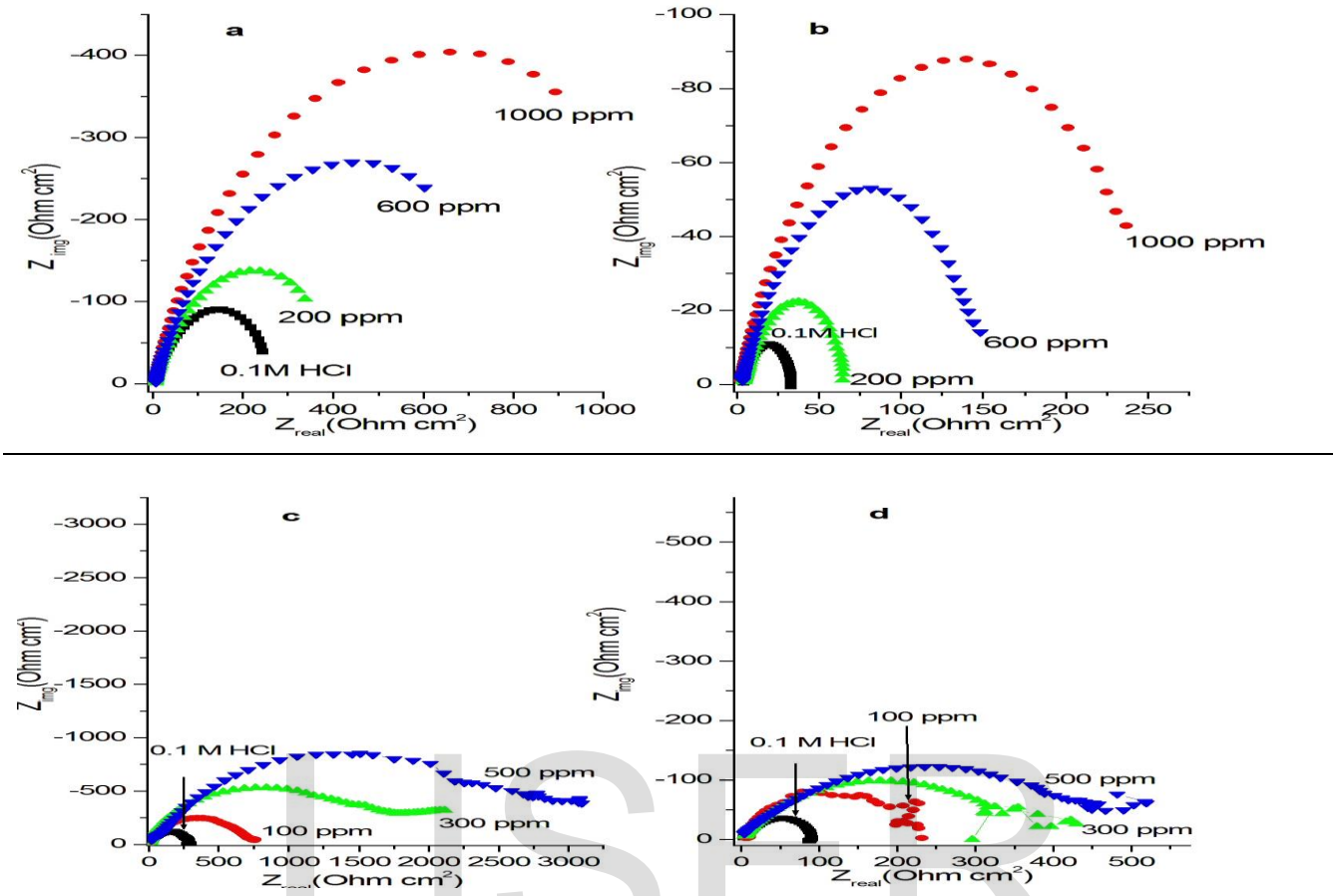
Electrochemical impedance spectroscopy is a non destructive quick method to analyze the interactions of corrosive atmosphere with the metal surface. Nyquist plots were recorded for zinc metal without and with different concentration of seroquel drug intermediate were shown in Fig. 4a and b and that for clotrimazole were shown in Fig. 4c and d respectively. The experimental curves obtained at room temperature and at elevated temperature were fitted to electrical equivalent circuits using ZSimpWin 3.21 and parameters computed were listed in Table 2 and 3 respectively.

Inhi.	C_{inh} (ppm)	R_p Ω cm²	C_{dl} F/cm²	Goodness of fit (x 10^{-4})	%η_z	θ
Seroquel	Blank	29.68	0.02129	4.172	-	-
	200	57.95	0.02187	4.863	48.78	0.49
	600	169.4	0.01401	3.625	82.47	0.82
	1000	213.9	0.00739	2.115	86.12	0.86
Clotrimazole	Blank	98.54	0.01135	1.3160	-	-
	100	225.64	0.01053	6.1290	56.32	0.56
	300	387.50	0.00389	4.0610	74.57	0.74
	500	523.94	0.00299	1.3250	81.19	0.81

Table 3 EIS studies of zinc metal in 0.1 M HCl solution without and with seroquel and clotrimazole drug intermediates at 333 K and 328 K respectively.

Results showed an increasing R_p and decreasing C_{dl} values in presence of increasing concentrations of the drug molecules at both the room temperature and elevated temperature. The inhibition efficiencies and surface coverage values were also found to increase with increasing concentrations of drug molecules at both the room temperature and elevated temperature. An increased diameter of Nyquist plot was seen with increased concentrations in case of both the drug intermediates. Nyquist plots for both inhibitors showed depressed semicircles.

Increased R_p and decreased C_{dl} values indicates the formation of a protective film in both the cases. Higher corrosion inhibition efficiency shown by seroquel at elevated temperature indicates involvement of chemical interactions between the metal surface and drug intermediate. But lower corrosion inhibition efficiency in case of clotrimazole at elevated temperature indicates involvement of physical adsorption. Depressed semicircles seen in Nyquist plots may be because of uneven surface of zinc metal samples [8, 9]. At ambient temperature clotrimazole showed a higher corrosion inhibition efficiencies compared to the seroquel. While seroquel showed a higher corrosion inhibition efficiencies compared to the clotrimazole



at elevated temperature.

Fig. 4 Nyquist plots for zinc in the absence and presence of different concentrations of **a)** seroquel at 303 K, **b)** seroquel at 333 K, **c)** clotrimazole at 298 K and **d)** clotrimazole at 328K.

Hence these results indicate, at ambient temperature clotrimazole while at elevated temperature seroquel is a better corrosion inhibitor for zinc in 0.1 M HCl solution.

3.3 Tafel polarization studies

It is one of the most commonly used electrochemical methods to know the rate of corrosion reactions. The Tafel polarization graphs obtained for both the drug intermediates are shown in Fig. 5. The data obtained by Tafel extrapolation were recorded in Table 4 and at elevated temperature was tabulated in Table 5.

Results showed that corrosion current decreased and so was the rate of corrosion reaction at both ambient and elevated temperature. Also the changes in corrosion potential values are less than 85 mV. A small change in both anodic and cathodic slopes of clotrimazole drug intermediates, but a little more change in anodic slope of seroquel drug intermediates compared to its cathodic slopes could be seen from the results.

Decreased corrosion current and rate is a clear indication of both the drug intermediates acting as efficient inhibitors. Clotrimazole may be acting as mixed type inhibitor as can be seen by Tafel slope values and by looking change in corrosion potential values [10]. By looking at the change in corrosion potential values and Tafel slope values it can be safe to say that the seroquel also acts as mixed type of inhibitor but with a more efficient in inhibiting anodic reactions.

$$\frac{C_{inh}}{\theta} = \frac{1}{K_{ads}} + C_{inh} \quad -----(8)$$

3.4 Adsorption isotherm

The inhibitor molecules may adsorb on the metal surface either through chemical or physical adsorption and thereby retarding the rate of corrosion. Different adsorption isotherms such Langmuir, Frumkin, Temkin and Freundlich isotherms were tested to know the mechanism of inhibitor adsorption [11, 12]. Surface coverage (θ) values obtained by EIS study were used for the adsorption isotherm estimation.

Seroquel adsorption on zinc surface followed Temkin while clotrimazole followed Langmuirs' adsorption isotherm model as shown in Fig. 6a and b respectively.

$$\theta = \frac{1}{f} \ln K_{ads} C_{inh} \quad -----(7)$$

The Temkin and Langmuirs' adsorption isotherms were expressed in terms of Eqs. (7) & (8).

Where K_{ads} is the adsorption equilibrium constant. The equilibrium constants K_{ads} are related to the standard adsorption free energy (ΔG^o) as shown in the Eq. (9).

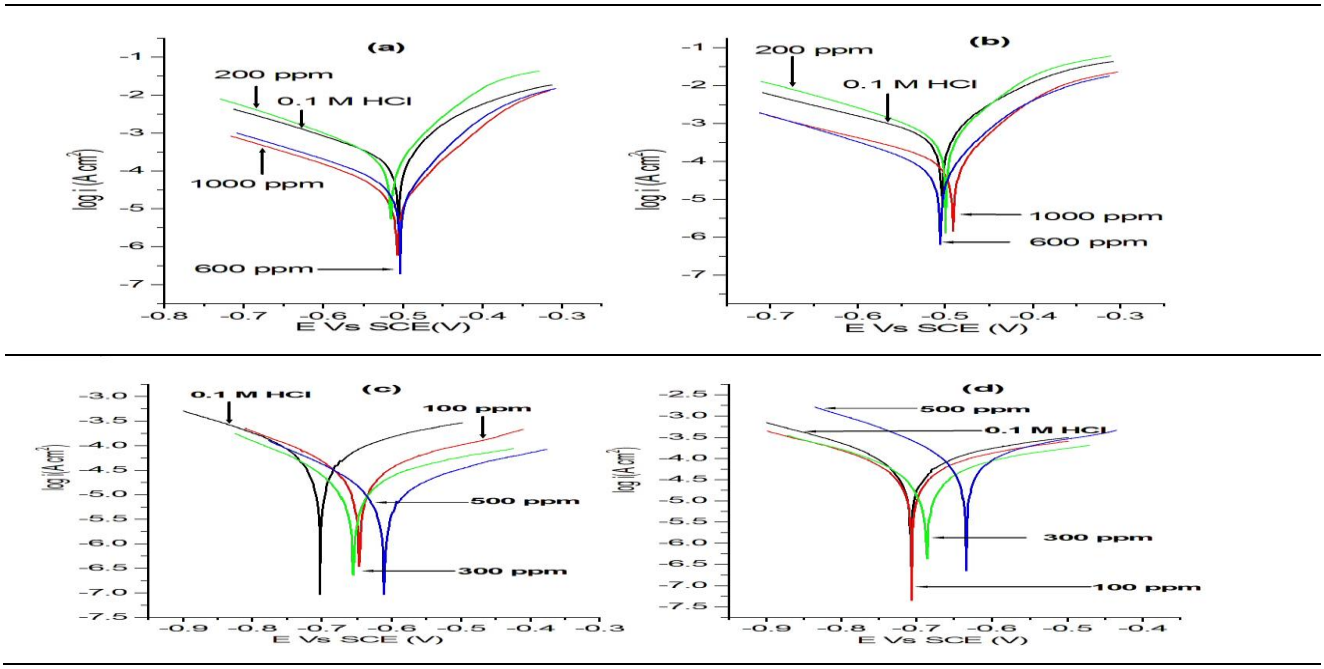


Fig. 5 Tafel polarization plots for zinc in the absence and presence different concentrations of **a)** seroquel at 303 K, **b)** seroquel at 333 K, **c)** clotrimazole at 298 K and **d)** clotrimazole at 328 K

Inhibitor	C_{inh} (ppm)	E_{corr} (V)	i_{corr} (A cm^{-2})	v_{corr} (mpy)	β_c mV/decade	β_a mV/decade	$\%\eta_p$	θ
Seroquel	Blank	-0.506	0.054	18.06	-7.008	09.648	-	-
	200	-0.515	0.039	12.80	-7.851	14.792	27.77	0.27
	600	-0.504	0.025	07.55	-7.040	16.183	53.70	0.53
	1000	-0.508	0.016	03.80	-7.228	17.381	70.37	0.70
Clotrimazole	Blank	-0.702	0.0458	10.91	5.569	4.507	-	-
	100	-0.647	0.0226	07.90	6.157	4.790	50.65	0.50
	300	-0.655	0.0131	04.59	6.707	4.102	71.39	0.71
	500	-0.611	0.0090	02.83	6.949	4.729	80.34	0.80

Table 4 Tafel polarization studies of zinc metal in 0.1 M HCl solution without and with seroquel and clotrimazole drug intermediates at room temperature

Inhibitor	C_{inh} (ppm)	E_{corr} (V)	i_{corr} (A cm^{-2})	v_{corr} (mpy)	β_c mV/decade	β_a mV/decade	$\%\eta_p$	θ
Seroquel	Blank	-0.503	0.130	31.00	-5.714	10.620	-	-
	200	-0.499	0.107	25.46	-7.301	12.671	17.69	0.17
	600	-0.505	0.052	15.70	-7.336	13.426	60.00	0.60
	1000	-0.491	0.023	09.02	-5.716	14.111	82.30	0.82
Clotrimazole	Blank	-0.707	0.0605	17.08	6.053	4.244	-	-
	100	-0.706	0.0431	14.40	5.793	4.460	28.76	0.28
	300	-0.686	0.0279	09.48	6.230	4.530	53.88	0.53
	500	-0.644	0.0195	04.99	6.854	4.048	67.76	0.67

Table 5 Tafel polarization studies of zinc metal in 0.1 M HCl solution without and with seroquel and clotrimazole drug intermediates at 333 K and 328 K respectively

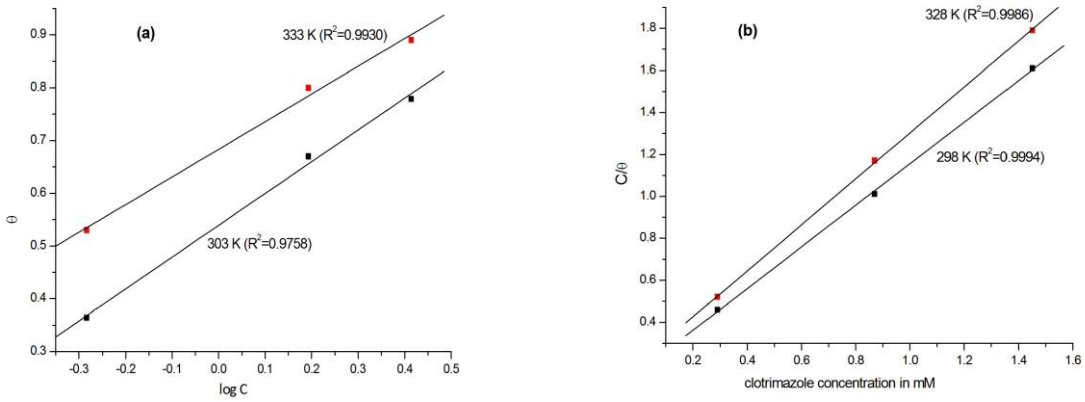


Fig. 6 a) Temkin adsorption isotherm for coverage of seroquel molecules on zinc surface **b)** Langmuir adsorption isotherm plot for coverage of clotrimazole molecules on zinc

	Temp.	K_{ads}	$-\Delta G^{O}_{ads}$ (kJmol^{-1})
Seroquel	303 K	1931	29.17
	333 K	1442	31.25
Clotrimazole	298 K	6172	31.57
	328 K	5405	34.39

Table 6 Adsorption parameters for inhibition of zinc surface corrosion by seroquel and clotrimazole drug intermediates in 0.1 M HCl solution

$$K_{ads} = \frac{1}{55.5} e^{\left(\frac{-\Delta G^{o}_{ads}}{RT}\right)} \quad -----(9)$$

Where 55.5 (molL^{-1}) is molar concentration of water in solution, T (K) is the absolute temperature and R (Jmol^{-1}K^{-1}) is the molar gas constant.

Adsorption isotherm parameters for both the drug intermediates were tabulated in Table 6. The standard free energy of adsorption (ΔG^{o}) values obtained in both the cases are found be and between -20 to -40 kJmol^{-1} and K_{ads} values were found to be high in both the drug intermediate.

It is generally accepted that, for values of $\Delta G°_{ads}$ up to −20 kJmol^{-1}, the type of adsorption can be regarded as physisorption and for values above −40 kJmol^{-1}; the adsorption is regarded as chemisorption [13, 14]. In case of both the drug intermediates $\Delta G°_{ads}$ values suggest the adsorption on zinc surface taking place by both the physisorption and chemisorption. High value of K_{ads} and negative $\Delta G°_{ads}$ suggest a spontaneous adsorption reaction on the metal surface [15] resulting in diminishing the rate of corrosion reaction.

3.5 Scanning electron microscopy

SEM images of zinc samples immersed in only 0.1 M HCl solution and in presence of optimum concentration of seroquel was shown in Fig. 7a and b respectively, while that of clotrimazole drug intermediate was shown in Fig. 8a and b respectively. It is clear by looking the micrographs that the zinc surface in presence of drug intermediate was smooth and less corroded compared to their absence in corrosive medium. It further confirms the formation of an adsorptive film on the zinc surface and thereby mitigating corrosion.

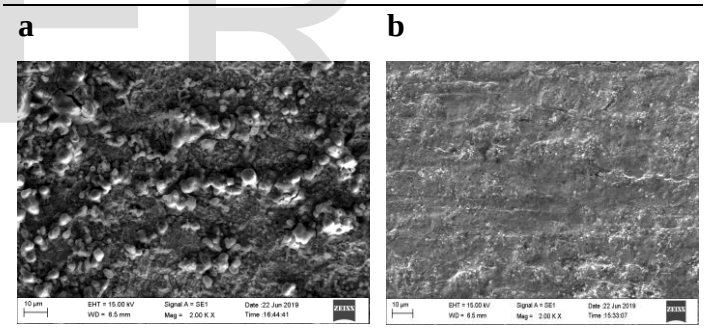


Fig.7 SEM micrograph of zinc sample surface immersed in 0.1 M HCl **a)** in the absence of seroquel inhibitor **b)**in the presence of 1000 ppm of seroquel inhibitor

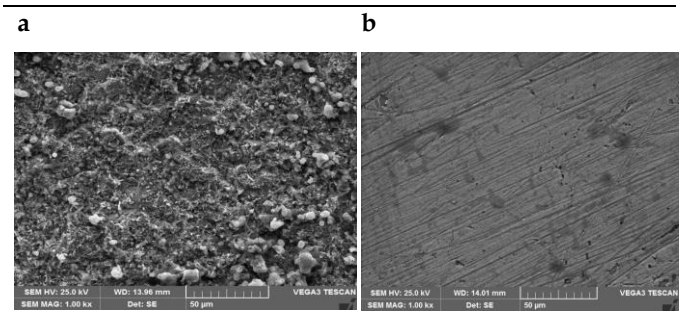


Fig. 8 SEM images for **a)** zinc corrosion in 0.1 M HCl in the absence of clotrimazole **b)** zinc corrosion in 0.1 M HCl in the presence of 500 ppm clotrimazole

4 Conclusions

The results obtained for both the drug intermediates by electrochemical studies were on par with that of mass loss method. The corrosion inhibition efficiency increased with inhibitor concentration and also with elevated temperature for seroquel. The seroquel showed a highest inhibition efficiency of 86.12% at 333 K as reported by EIS studies. The corrosion inhibition efficiency increased with inhibitor concentration but decreased at elevated temperature for clotrimazole. The clotrimazole showed a highest inhibition efficiency of 90% at ambient temperature as reported by EIS studies. The results from Tafel polarization studies for both the drug intermediates indicated the inhibition of anodic and cathodic corrosion reactions. The seroquel was found to fallow Temkin adsorption isotherm while clotrimazole Langmuirs'. The higher values of K_{ads} and negative values of ΔG^{o}_{ads} in case of both the drug intermediate adsorption indicated a spontaneous adsorption process. SEM images also confirmed the decreased unevenness on the zinc surface in presence of both drug intermediates compared to their absence. It can be conclude that corrosion inhibition efficiency of seroquel is more at studied elevated temperature compared to ambient temperature. Whereas corrosion inhibition efficiency of clotrimazole is more at ambient temperature compared to studied elevated temperature.

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