

A Study of Aluminium Corrosion Inhibition in Acid Medium by an Antiemetic Drug

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Received: 7 January 2011 / Accepted: 11 May 2011 / Published online: 1 December 2011
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Abstract The inhibition effect of meclizine hydrochloride on the corrosion behaviour of aluminium in 1 M hydrochloric acid medium was studied by weight loss, potentiodynamic polarisation and electrochemical impedance spectroscopy techniques. Results obtained revealed that Meclizine performed excellently as a corrosion inhibitor for aluminium in HCl medium at 303 K. The effect of temperature on inhibition efficiency was studied. The inhibition was assumed to occur via adsorption of the inhibitor molecule on the metal surface. The adsorption of the inhibitor on the aluminium surface followed Langmuir adsorption isotherm model. The values of activation energy and thermodynamic parameters were calculated and discussed.

Keywords Aluminium · Acid corrosion · Weight loss · EIS · Polarisation

1 Introduction

One of today's most important considerations in industry is reduction of overall costs by protection and maintenance of materials used. The protection of corroding metal surface prevents the waste of both resources and money during the industrial applications and is vital for the extension of lifetime of the equipment and limiting the dissolution of

the toxic metals from the components into the environment.

Aluminium and its alloys find a wide spectrum of technological applications due to their particular properties such as low density, good appearance and corrosion resistance. However aluminium and its alloys are prone to undergo corrosion in aggressive acidic media due to the dissolution of the passive film. Acid solutions are widely used for pickling, chemical and electrochemical etching of aluminium. The presence of aggressive anions like chloride creates extensive localised attack. Due to general aggressiveness of the acid solutions corrosion inhibitors are commonly used to reduce the corrosion attack on metal surfaces.

The use of specific chemical compound as an inhibitor is the simple, practical and cost effective means of preventing the corrosion of metals. The corrosion inhibitors bring down the rate of corrosion to a greater extent, even when added in small quantities to the corrosive environment. Corrosion inhibitors used can be organic or inorganic compounds. To act as an inhibitor inorganic compound must be able to oxidize the metal forming passive layer, and concentration of the inhibitor must be above a critical concentration, failing which metal to be protected would suffer from highly localized corrosion. More over many inorganic compounds like nitrites [1], chromates [2], arsenic salts [3], which exert powerful inhibiting effect, have limited application due to their toxicity while organic compounds with N, S, O, atoms or π -electrons easily get adsorbed on the metal surface retarding corrosion. A large number of organic compounds were studied as corrosion inhibitors [4–13]. Though many organic compounds show good anticorrosive ability, most of them are highly toxic, very expensive and environmentally hazardous. Due to increasing environmental awareness and adverse effect of

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some chemicals, developing cheap, non-toxic and environmentally benign corrosion inhibitors are of great importance. Recently use of drugs as corrosion inhibitor for metals in various aggressive media has generated lot of interest. The research in this area will serve as a possible replacement for toxic and costly organic inhibitors.

Many investigators have reported the use of antibacterial [14–19], antifungal [20, 21], antibiotics [22–24], sulpham [25] and azo-sulpha drugs [26] as corrosion inhibitors. Meclizine hydrochloride is an antiemetic oral drug, used as antihistamine to treat or prevent motion sickness, nausea and vomiting associated with a variety of conditions [27–29]. Meclizine is approved by the U.S. Food and Drug Administration (FDA). It is a piperazine derivative, containing N-atoms and π -bonds in its structure which is regarded as one of the important factors to be a good inhibitor. Inhibitor molecule is big enough to efficiently cover more surface area of the metal. Besides, it is cheap, easily available, non toxic and environment friendly. The aim of the present investigation is to use meclizine hydrochloride, which is a non-toxic antiemetic drug, as a potential inhibitor for corrosion of aluminium in 1 M HCl.

2 Experimental

The test materials used for corrosion measurements were aluminium samples with chemical composition, wt%: 0.1Cu, 0.4–0.9Mg, 0.3–0.7Si, 0.6Fe, 0.3Mn, 0.2Zn, 0.2Cr, 0.1Ti/other graining elements and remainder Al. Test materials were polished with different emery papers up to 1,000 grade, cleaned with acetone, washed with double distilled water and properly dried prior to exposure. Analar grade HCl and double distilled water were used to prepare all solutions. Meclizine hydrochloride (F.W.481.885) from Frapps Chemical Reagent Co. Ltd was used for the study. The IUPAC name of the compound is 1-(*p*-chloro- α -phenylbenzyl)-4-(*m*-methylbenzyl) piperazine dihydrochloride monohydrate. Figure 1 shows the molecular structure of meclizine hydrochloride.

Weight loss measurements were performed on aluminium coupons having $1 \times 0.5 \times 0.06$ inch size in 1 M

hydrochloric acid solution with different concentrations of the inhibitor. Weight loss of the metal coupons was noted after an immersing period of 4 h at the temperature range from 303 to 333 K.

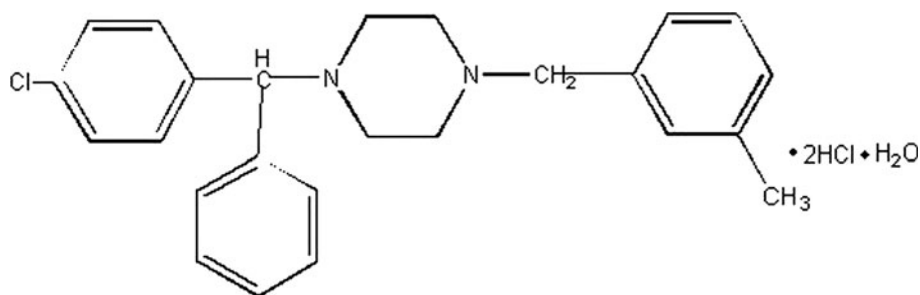
Electrochemical tests were carried out with a CH-analyser model-CH1660D. The cell arrangement used was a conventional three electrode cell with platinum counter electrode, saturated calomel electrode as reference electrode and test material (Al) as working electrode. The test material was covered by epoxy adhesive-araldite, so that only 1 cm^2 area was in contact with the solution. Polarisation curves were recorded, potentiodynamically at the scan rate of 1 mV/s, in the range of +250 mV to –250 mV versus Open Circuit Potential (OCP). Impedance was measured over a frequency range of 1,00,000–0.05 Hz using an amplitude of 10 mV peak to peak using AC signal. The surface of aluminium was analysed after the corrosion tests, by an scanning electron microscope.

3 Results and Discussion

3.1 Polarisation Measurements

Figure 2 shows the potentiodynamic polarisation curves for aluminium in 1 M HCl containing 0, 10, 50, 100, 200 ppm of the inhibitor at 303 K. Corrosion current (i_{corr}) was found to decrease with increase in inhibitor concentration indicating, the increased inhibition efficiency with the increase in the concentration of the inhibitor. In acidic solution the anodic process of corrosion is the passage of metal ions from the solid metal to the solution, and the principal cathodic process is the discharge of hydrogen ions to produce hydrogen molecules or reduction of oxygen. The inhibitor may affect either of them or both anodic and cathodic process. In the present study, presence of inhibitor resulted in marked shift in the cathodic and anodic branches of the polarisation curves. Moreover, in the presence of inhibitor the values of corrosion potential E_{corr} are nearly constant; therefore, meclizine hydrochloride could be classified as a mixed type inhibitor. The values associated with electrochemical parameter and calculated

Fig. 1 Molecular structure of meclizine hydrochloride



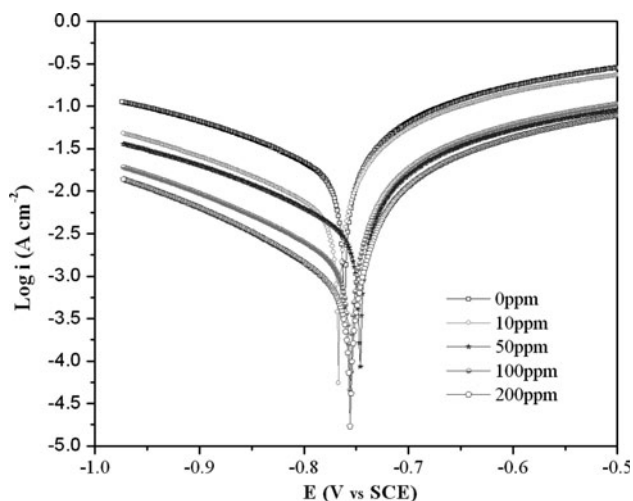


Fig. 2 Potentiodynamic polarisation curves of Al in 1M HCl solution at 303 K containing various concentrations of inhibitor

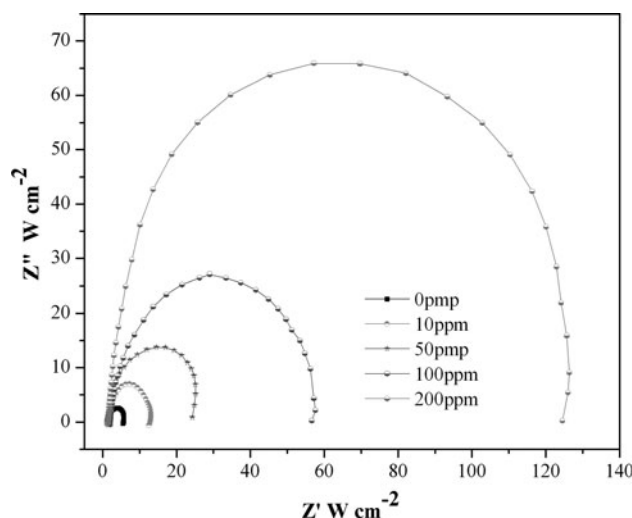


Fig. 3 Nyquist plots for Al in 1 M HCl solution at 303 K containing various concentrations of inhibitor

inhibition efficiency (IE %) are given in Table 1. The inhibition efficiency is given by Eq. 1 [30]:

$$IE (\%) = \left(1 - \frac{i'_{corr}}{i_{corr}} \right) \times 100 \tag{1}$$

where, i'_{corr} and i_{corr} are the corrosion currents in the presence and absence of the inhibitor respectively.

3.2 Electrochemical Impedance Spectroscopic Measurements (EIS)

The impedance spectra (Nyquist plots) of aluminium in 1 M HCl containing 0, 10, 50, 100, 200 ppm of the inhibitor at 303 K is shown in Fig. 3. Nyquist plots contain depressed semicircle with centre under real axis. The size of the semicircle increases with the inhibitor concentration, indicating the charge transfer process as the main controlling factor of the corrosion of aluminium. It is apparent from the plots that, the impedance of the inhibited solution has increased with the increase in the concentration of the inhibitor. The experimental results of EIS measurements for the corrosion of aluminium in 1 M HCl in the absence and presence of inhibitor is given in Table 2. It can be observed that polarisation resistance (R_p) value increased with increase in the concentration of the inhibitor. Whereas

Table 2 AC impedance data of Al in 1 M HCl solution at 303 K

[Inhibitor]/ppm	R_p	IE (%)	C_{dl}
0	5.74	–	88.72
10	13.09	56.09	83.04
50	25.22	77.21	56.32
100	57.38	89.98	34.1
200	125.68	95.4	22.05

values of the capacitance of the interface (C_{dl}) starts decreasing, with increase in inhibitor concentration, which is most probably due to the decrease in local dielectric constant and/or increase in thickness of the electrical double layer. This suggests that the inhibitor acts via adsorption at the metal/solution interface [31] and the decrease in the C_{dl} values is caused by the gradual replacement of water molecules by the adsorption of the inhibitor molecules on the electrode surface, which decreases the extent of metal dissolution [32].

The inhibition efficiency is given by Eq. 2 [33]:

$$\%IE (R_p) = \frac{R_{p(inhi)} - R_p}{R_{p(inhi)}} \times 100 \tag{2}$$

Table 1 Electrochemical parameters of Al in 1 M HCl solution at 303 K

[Inhibitor]/ppm	$i_{corr} \times 10^{-3}$ (A)	CR $\times 10^3$ (mil/year)	E_{corr} (V)	β_a (V/dec)	β_c (V/dec)	IE (%)
0	41.97	17.99	–0.76	5.082	4.625	–
10	18.68	8.007	–0.7681	5.448	4.971	55.49
50	10.12	4.339	–0.746	5.288	5.320	76.06
100	4.57	1.958	–0.7544	5.814	5.667	88.93
200	1.933	0.884	–0.7556	6.383	6.428	95.39

Table 3 Corrosion parameters for aluminium in 1 M HCl in the absence and presence of different concentrations of the inhibitor at 303 K

[inhibitor]/ppm	Weight loss (mg cm^{-2})	IE (%)
0	24.23	–
5	14.14	41.69
10	9.79	59.6
50	5.64	76.66
100	2.76	88.55
200	1.837	92.34
500	1.71	92.9

where R_p is polarisation resistance without inhibitor, and $R_{p(\text{inhi})}$ is polarisation resistance with inhibitor.

C_{dl} value is obtained from the Eq. 3:

$$\omega_{\max} = 2\pi f_{\max} = \frac{1}{R_p C_{dl}} \quad (3)$$

where, f_{\max} is the frequency at the top of the semicircle (where $-Z''$ is maximum).

3.3 Weight Loss Measurements

3.3.1 Effect of Inhibitor Concentration

The weight loss results of aluminium in 1 M HCl in the absence and presence of various concentrations of the inhibitor are summarized in Table 3. The IE was calculated using the following Eq. 4.

$$\text{IE (\%)} = \frac{W_2 - W_1}{W_2} \times 100 \quad (4)$$

where W_1 and W_2 are weight loss of aluminium in the presence and absence of the inhibitor.

The inhibition efficiency increased with increase in the concentration of the inhibitor. Results obtained from polarisation, EIS and weight loss measurements are in good agreement with each other. Reported drug Meclizine.HCl has better inhibition efficiency than the already reported drugs [21, 25].

3.3.2 Effect of Temperature

To elucidate the mechanism of inhibition and to determine the thermodynamic parameters of the corrosion process weight loss measurements were performed at 303, 313, 323, 333 K. The effect of temperature on the corrosion inhibition of aluminium in the presence of the inhibitor is graphically represented in Fig. 4.

The inhibition efficiency decreased with increase in temperature, leading to the conclusion that the protective

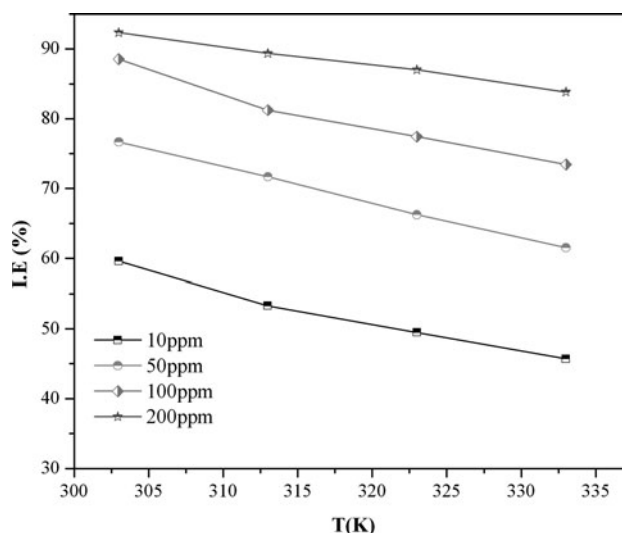


Fig. 4 Effect of temperature on IE for aluminium in 1 M HCl in the presence of different concentrations of the inhibitor

film of these compounds formed on the aluminium surface is less stable at higher temperature; which may be due to the desorption of some adsorbed molecules from the surface of the aluminium at higher temperature due to which greater area of the metal is exposed to the acidic environment.

The apparent activation energy (E_a°) of metal corrosion in acid media can be calculated from the Arrhenius equation [34].

$$\ln CR = \frac{E_a^\circ}{RT} + A \quad (5)$$

where, E_a° is the apparent activation energy for the corrosion of aluminium, R is the universal gas constant, A is Arrhenius pre exponential factor and T is the absolute temperature. Figure 5 depicts the plot of $\log CR$ versus $1/T$ and the values of E_a° obtained from the slope of the plot are given in Table 4.

The relationship between the temperature dependence of inhibition efficiency of an inhibitor and the activation energy found in its presence was given as follows [35]:

- Inhibitors whose IE decreases with temperature increase. The value of activation energy (E_a°) found is greater than that in the uninhibited solution.
- Inhibitors whose IE does not change with temperature variation. The value of activation energy (E_a°) does not change with the presence or absence of inhibitors.
- Inhibitors whose IE increases with temperature increase. The value of activation energy (E_a°) found is less than that in the uninhibited solution.

The higher value of activation energy (E_a) in the presence of inhibitor than in its absence is attributed to its

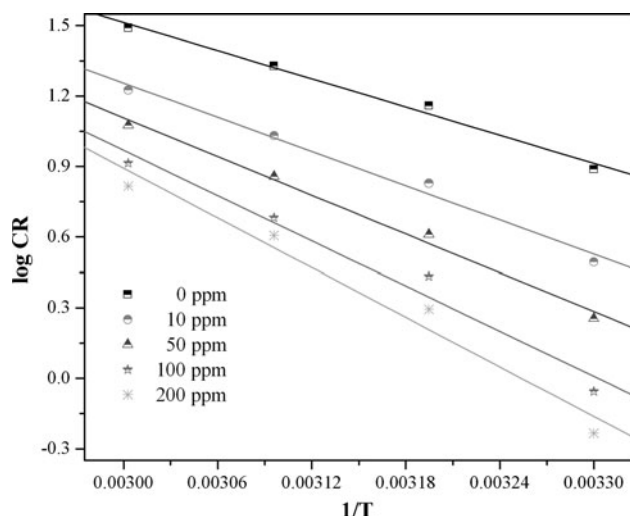


Fig. 5 Arrhenius plot for aluminium in 1 M HCl solution in the absence and presence of different concentrations of the inhibitor

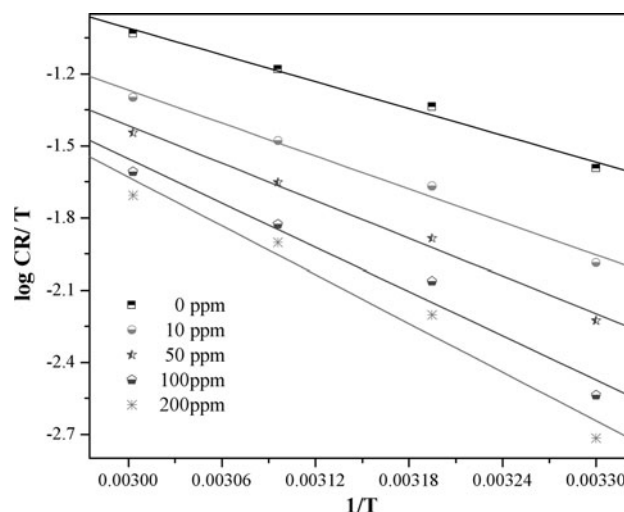


Fig. 6 Transition state plot for aluminium in 1 M HCl solution in the absence and presence of different concentrations of the inhibitor

Table 4 Values of activation parameters E_a° , ΔS_a° , ΔH_a° for aluminium in 1 M HCl in the absence and presence of different concentrations of the inhibitor

[Inhibitor]/ppm	E_a° (kJ mol ⁻¹)	ΔH_a° (kJ mol ⁻¹)	ΔS_a° (J mol ⁻¹)
0	38.32	35.68	-109.8
10	46.47	43.83	-90.3
50	52.52	49.88	-74.9
100	61.35	58.71	-51.1
200	67.32	64.68	-34.6

physical adsorption, its chemisorption is pronounced in the opposite case [21].

In the present study the higher value of E_a° for aluminium in an inhibitor’s presence, compared to that in its absence is attributed to its physical adsorption. Szauer and Brand [36] explained that the increase in E_a° can be attributed to an appreciable decrease in the adsorption of the inhibitor on the aluminium surface with increase in temperature and a corresponding increase in corrosion rates occurs due to the fact that greater area of metal is exposed to acid environment. An alternative form of Arrhenius equation is the transition state equation [31]

$$CR = \frac{RT}{Nh} \exp\left(\frac{\Delta S_a^\circ}{R}\right) \exp\left(-\frac{\Delta H_a^\circ}{RT}\right) \quad (6)$$

where, h is the Plank’s constant, N is the Avogadro’s number, ΔS_a° is the entropy of activation, and ΔH_a° is the enthalpy of activation. A plot of $\log (CR/T)$ versus $1/T$ gave a straight line as shown in Fig. 6 with a slope of $(-\Delta H_a^\circ/2.303R)$ and an intercept of $[\log(R/Nh) + (\Delta S_a^\circ/R)]$, from which the values of ΔH_a° and ΔS_a° were calculated and listed in Table 4.

The positive values of ΔH_a° for corrosion of aluminium in the presence and absence of the inhibitor reflect the endothermic nature of metal dissolution process. The increase in ΔH_a° with increase in the concentration of the inhibitor for aluminium corrosion reveals that decrease in aluminium corrosion rate is mainly controlled by kinetic parameters of activation [37].

The increase in ΔS_a° with increase in the concentration of the inhibitor for aluminium corrosion reveals that randomness is increased on going from reactant to the activated complex [38].

3.3.3 Adsorption Isotherms

In order to understand the mechanism of corrosion inhibition, the adsorption behaviour of the organic adsorbate on the metal surface has to be known. The degree of surface coverage (Θ) for different concentration of the inhibitor (C_{inh}) has been evaluated. The data were tested graphically by fitting to various isotherms. A straight line with correlation coefficient nearly equal to 1.0 was obtained on plotting C_{inh}/Θ against C_{inh} as shown in Fig. 7 suggesting, adsorption of the compound on the aluminium surface followed Langmuir adsorption isotherm model. It is interpreted from the graph that, adsorption of the inhibitor on the aluminium surface followed Langmuir adsorption isotherm at all studied temperatures. According to this isotherm, the surface coverage (Θ) is related to inhibitor concentration C_{inh} [39] by Eq. 7,

$$\frac{C_{inh}}{\theta} = \frac{1}{K_{ads}} + C. \quad (7)$$

where, K_{ads} is equilibrium constant of the equilibrium adsorption process. This isotherm assumes that adsorbed

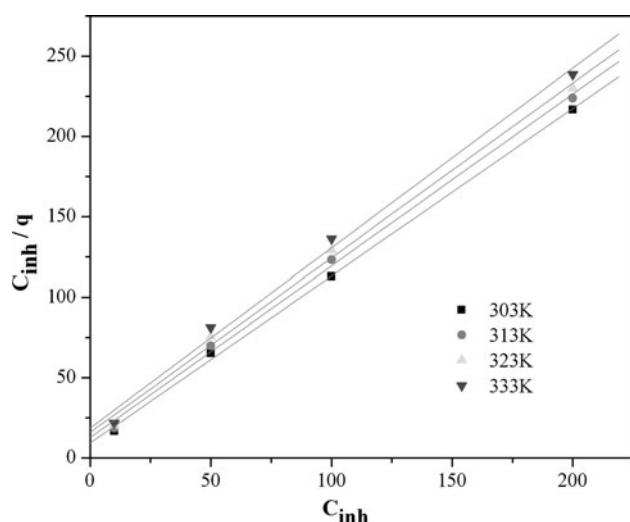


Fig. 7 Langmuir adsorption isotherm for aluminium in 1 M HCl solution at various temperatures

molecule occupies only one site and it does not interact with other adsorbed species.

The K_{ads} values can be calculated from the intercept lines on the C_{inh}/Θ axis. This is related to the standard free energy of adsorption ($\Delta G_{\text{ads}}^{\circ}$) by Eq. 8:

$$\Delta G_{\text{ads}}^{\circ} = -RT \ln (55.5K_{\text{ads}}) \quad (8)$$

where, R is the gas constant and T is the absolute temperature. The constant value of 55.5 is the concentration of water in solution in mol/l. The values of $\Delta G_{\text{ads}}^{\circ}$ for the inhibitor on the surface of aluminium are given in Table 5.

The negative value of G_{ads}° indicated spontaneous adsorption of the inhibitor on the aluminium surface. Generally, the magnitude of $\Delta G_{\text{ads}}^{\circ}$ around -20 kJ/mol or less negative indicates electrostatic interactions between inhibitor and the charged metal surface (i.e., physisorption). Those around -40 kJ/mol or more negative are indicative of charge sharing or transferring from organic species to the metal surface to form a coordinate type of metal bond (i.e., chemisorptions). In the present work, the calculated values $\Delta G_{\text{ads}}^{\circ}$ at 303 K for aluminium is -38 kJ/mol, which indicate that adsorption of the inhibitor on the aluminium surface involves both physical and chemical process [40–42]. But the inhibition efficiency decreased

Table 5 Values of thermodynamic parameters for the adsorption of the inhibitor in 1 M HCl on the aluminium at different temperatures

Temperature (K)	K (mol ⁻¹)	$\Delta G_{\text{ads}}^{\circ}$ (kJ mol ⁻¹)	$\Delta H_{\text{ads}}^{\circ}$ (kJ mol ⁻¹)	$\Delta S_{\text{ads}}^{\circ}$ (J mol ⁻¹)
303	51,805	-37.46	-19.58	59.03
313	37,648	-37.87	-19.58	58.44
323	30,265	-38.49	-19.58	58.56
333	25,684	-39.23	-19.58	59.02

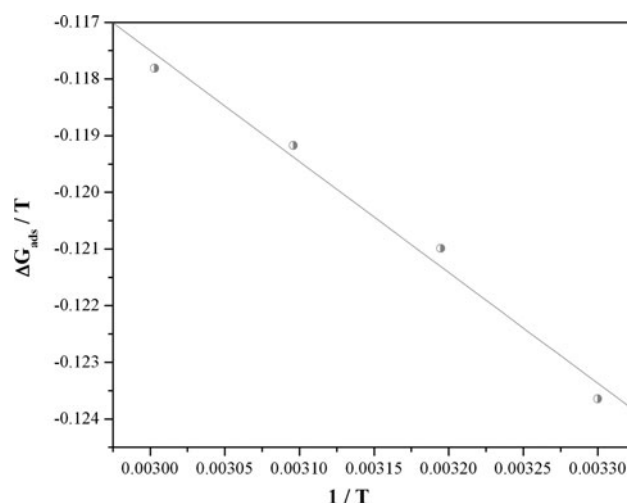


Fig. 8 $\Delta G_{\text{ads}}^{\circ}/T$ versus $1/T$ plot for aluminium in 1 M HCl solution

with increasing temperature indicating inhibitor adsorbed predominantly physically on the surface of aluminium. The enthalpy of adsorption ($\Delta H_{\text{ads}}^{\circ}$) can be calculated from the rearranged Gibbs–Helmholtz equation:

$$\frac{\Delta G_{\text{ads}}^{\circ}}{T} = \frac{\Delta H_{\text{ads}}^{\circ}}{T} + K \quad (9)$$

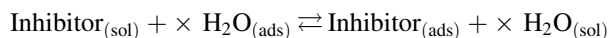
The variation of $\Delta G_{\text{ads}}^{\circ}/T$ with $1/T$ gave a straight line with a slope of $\Delta H_{\text{ads}}^{\circ}$ as shown in Fig. 8. The entropy of adsorption ($\Delta S_{\text{ads}}^{\circ}$) was calculated using the following thermodynamic equation:

$$\Delta S_{\text{ads}}^{\circ} = \frac{\Delta H_{\text{ads}}^{\circ} - \Delta G_{\text{ads}}^{\circ}}{T} \quad (10)$$

It is well known fact that adsorption is an exothermic phenomenon accompanied by a decrease in entropy [43]. In aqueous solutions the adsorption of the organic molecule is generally accompanied by desorption of water molecules [29]. The calculated values of heat of adsorption and entropy of adsorption are listed in Table 5. The negative sign of $\Delta H_{\text{ads}}^{\circ}$ indicated the exothermic process of adsorption of the inhibitor on aluminium surface in HCl. The positive value of $\Delta S_{\text{ads}}^{\circ}$ in the presence of inhibitor can be attributed to the increase in the solvent entropy and more positive desorption entropy. It is also interpreted that the increase of disorders is due to more water molecules which can be desorbed from the metal surface by one inhibitor molecule. Therefore it is revealed that decrease in the enthalpy is the driving force for the adsorption of the inhibitor on the surface of aluminium [6, 44, 45].

3.4 Mechanism of Corrosion Inhibition

The adsorption of the inhibitor molecule is often a displacement reaction involving removal of adsorbed water molecules from the metal surface.



In general the physisorption involves electrostatic force of attraction between electrically charged surface of metal and charged species in the bulk of this solution. While chemisorptions involve charge sharing or electrons transfer from the inhibitor molecule to the metal surface to form co-ordinate type of bond.

In hydrochloric acid medium, the metal surface is negatively charged due to the specifically adsorbed Cl^- ions on the metal surface. In acidic solution, the nitrogen atom of the piperazine ring of the inhibitor can be protonated easily, due to high electron density on it, leading to positively charged inhibitor species. The adsorption can occur via electrostatic interaction between positively charged inhibitor molecules and negatively charged metal surface [17, 32]. Further, co-ordinate bond may be formed between unshared e^- pairs of unprotonated nitrogen atom of the inhibitor and metal surface. Additionally inhibitor molecule may be chemically adsorbed due to interaction of π electrons of the inhibitor with metal surface [37].

3.5 Scanning Electron Microscopy

To evaluate the conditions of the metal surface in contact with acid solution in the absence and presence of inhibitor, Scanning Microscopic Analysis (SEM) was carried out, immediately after the corrosion tests. The aluminium samples in 1 M HCl solution with and without optimal concentration of the inhibitor were subjected to analysis. The SEM micrographs are shown in Fig. 9 revealed that; the surface corrosion of aluminium was decreased remarkably in the presence of the inhibitor.

4 Conclusion

- Meclizine hydrochloride is a good inhibitor for the corrosion of aluminium in 1 M HCl and inhibition efficiency was more pronounced with increase in the inhibitor concentration.
- The results obtained from polarisation, impedance techniques and weight loss measurements are in good agreement.
- The potentiodynamic polarisation curves imply that, meclizine hydrochloride acts as a mixed type inhibitor for corrosion of aluminium in 1 M HCl.
- The values of $\Delta G_{\text{ads}}^{\circ}$ indicate adsorption of the inhibitor by both physical and chemical process while the decrease in inhibition efficiency with increase in temperature indicated predominate physisorption of the inhibitor.
- The adsorption of inhibitor on the aluminium surface obeys Langmuir adsorption isotherm.

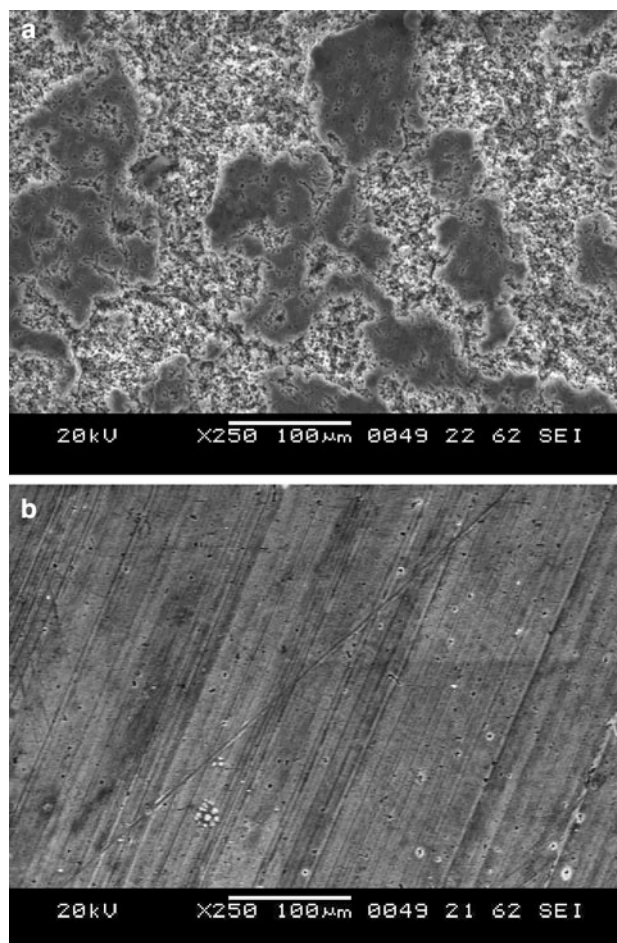


Fig. 9 SEM micrographs of surface of Al after immersion for 4 h at 303 K in **a** 1 M HCl, **b** 1 M HCl + 200 ppm inhibitor

Acknowledgment Authors thank Dr. Arthoba Naik and Dr. Basavanna of Kuvempu University for their help and support.

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