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Effect of Temperature of Calotropis Gigantea leaves on Aluminum Corrosion in Hydrochloric Acid

P. S. Desai[1]*

Department of Chemistry, Arts, Science and Commerce College,
Kamrej Char Rasta, Surat 394 185, Gujarat, INDIA.

ABSTRACT

The purpose of this paper to find out the effect of Calotropis Gigantea (Aankado) leaves ark's on the corrosion of aluminum in 0.5M hydrochloric acid (HCl) at varied temperatures 313K -333K, and elucidates the mechanism of the inhibition system. Gravimetric method was used to investigate the effect of aluminum corrosion in HCl solutions. The corrosion inhibitive effect of Calotropis Gigantea (Aankado) leaves ark's on aluminum in 0.5M HCl solution the inhibition efficiency have been carried out using gravimetric (weight loss) method at 313 to 333K. The trend of inhibition efficiency with temperature was used to suggest the mechanism of inhibition. The result explained that Calotropis Gigantea (Aankado) leaves ark's acts as an inhibitor for acid induced corrosion of aluminum. Inhibition efficiency was found to increase with extract concentration. On the other hand inhibition efficiency (η%) increased with the increase of inhibitor concentration but decreased with increase in temperature. Inhibition adsorptions characteristic were estimated by Langmuir and Freundlich adsorption isotherm at all concentration and temperature studied. The paper deal with the inhibition of corrosion of commercially pure aluminum alloy in 0.4, 0.5 and 0.6 molars of HCl.

***Keywords*: Adsorption, Aluminum, Calotropis Gigantea, Corrosion, Hydrochloric acid, Langmuir isotherm.**

I. INTRODUCTION

Aluminum not found free in nature. Aluminum is an abundant element in the earth's crust. Alloys with small amounts of copper, magnesium, silicon, manganese, and other elements have very useful properties. Strength depends on purity. Aluminum alloy is known to exhibit passive behavior in aqueous solutions. The corrosion of the metal has been reported to depend on processes associated with the passivating surface oxide film such as metal ion transfer to the metal/oxide interface, metal ion and oxygen ion transfer to the oxide/solution interface, ion migration in the oxide film, and electron transfer from the metal to acceptor species in solution [1].

Hydrochloric acid solutions are commonly used for pickling of aluminum and for its chemical and electrochemical etching of metal materials. The corrosion rates of metals or alloys can be covered up appreciable through surface modification by adsorbed organic molecules. The inhibiting effect is depending upon the adsorption ability of their molecules. The inhibitor after adsorption may form a surface film that acts as a physical barrier restricting the diffusion of ions/molecules to or from the metal/alloy surface and may prevent the metal atoms from participating in either the anodic or cathodic reactions of corrosion [2-3].

In recent years, attempts have been made to understand the nature of interaction between the inhibitor and metal surface in terms of adsorptions isotherm [4,5]. It was reported that there is specific adsorption of the inhibitors

IJARSE
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on the metal surface, the corrosion reaction proceeds by diffusion of the corrosive species, though fine pores of the protective layer formed.

Organic compounds are commonly used as corrosion inhibitors [6-8] but organic molecules are highly expensive and hazardous to environment. Therefore, it is desirable to source for environmentally safe inhibitors [9-14]. It has been shown that natural products of plant origin contain various organic compounds e.g. alkaloids, tannins, pigments, organic and amino acids, and most are known to have inhibitive action [15-22]. We have been investigated a number of potential eco-friendly inhibitors for mild steel corrosion in hydrochloric acid solution [23-25]. Therefore, in this regards, the present article reports on the inhibitive action of Calotropis Gigantea (Aankado) leaves ark's on the corrosion of aluminum alloy in 0.5M HCl solutions using gravimetric method at varied temperature 313 to 333K.

II. EXPERIMENTAL

2.1 Materials

Rectangular specimens of the size 5.0 x 2.0 x 0.18 cm having an area of 0.2259 sq. dm of 2S grade aluminum alloy (Al = 98.02 %; Mg = 0.37 %; Si = 0.49 %; Fe = 0.68 %; Mn = 0.16 % and Cu = 0.082 %.) with small hole of about 5 mm diameter near the upper edge, were used for the determination of the corrosion rate.

Ankado Leaves Extract: Take Calotropis Gigantea plant leaves were dried, grind to powder form and boiling with double distilled water to making extract of different concentrations 0.25, 0.5, 0.75, 1.0 and 1.25 %.

All chemicals and reagents used were of analytical grade and used as source without further purification. The aggressive media was 0.5M HCl solution. Inhibitor Calotropis Gigantea leaves ark's was used in the concentration range 0.25 to 1.25%.

2.2 Determination of Weight Loss

The procedure for weight loss determination was similar to that reported earlier [6]. According to this method [6], previously weighed aluminum specimens were immersed in 250 ml open beakers containing 230 ml of 0.5M HCl (blank) and then with addition of different concentrations of Calotropis Gigantea extract to the 0.5M HCl (0.25-1.25%) at 313-333K. The variation of weight loss was observed after 2 hours immersion per coupon at 313 to 333K temperatures respectively. After 2 hours, the coupons were taken out, immersed in Chromic-Phosphate [26] mixture at room temperature, scrubbed with a bristle brush under running water, dried and reweighed. The weight loss was calculated in milligram as the difference between the initial weight and the weight after the removal of the corrosion product. The experimental readings were recorded using contact digital analytical balance (CAH 123) electronic weighing balance with the accuracy of ±0.001gram. Triplicate experiments were performed for each concentration of inhibitor.

III. RESULTS AND DISCUSSION

The results are presented in Tables I to III and Figures 1 to 6. To assess their protective value, Calotropis Gigantea extract was added to solutions of hydrochloric acid.

IJARSE
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The corrosion rate was calculated from the weight loss data with help of equation (1). Where, 'W' is the weight loss of Aluminum in grams, 'A' is the surface area of specimen in inches square, 'D' is the density of aluminum and 't' is the time in hours.

$$CR\ (mpy) = \frac{534\,W}{DAt} \qquad (1)$$

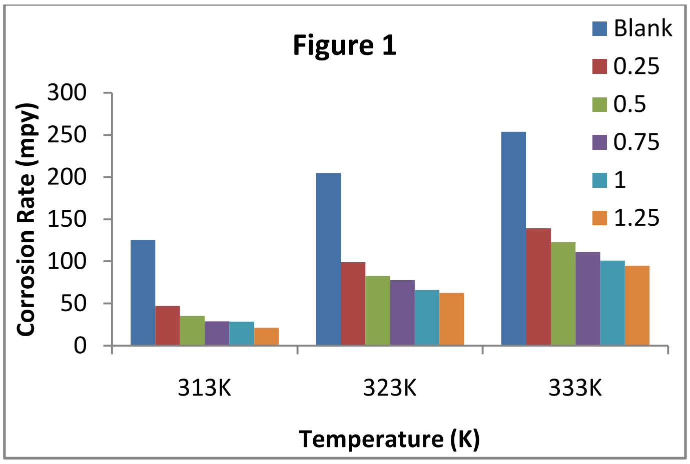

Figure 1: Corrosion rate of presence and absence inhibitor in 0.5M HCl at various

The rising of temperature, the corrosion rates are shown in Figure 1. The results suggest that increasing temperature there is an increase of corrosion rate in the absence and presence of Calotropis Gigantea extract. The increases of corrosion rate in the absence of extract is higher at all temperatures studied, revealed that the increase in corrosion rate with increase in temperature may be probably due to decreasing strength of adsorption and roughening of the electrode surface which results from enhanced corrosion [27].

The inhibition efficiency (η%) and degree of surface coverage (θ) at each concentration of ark of Calotropis Gigantea extract were calculated by comparing the corrosion rate in absence (CR_{blank}) and presence of inhibitor (CR_{inh}) using the relationships:

$$\eta\% = \left(\frac{CR_{blank} - CR_{inh}}{CR_{inh}}\right) \times 100 \qquad (2)$$

$$\theta = \left(\frac{CR_{blank} - CR_{inh}}{CR_{inh}}\right) \qquad (3)$$

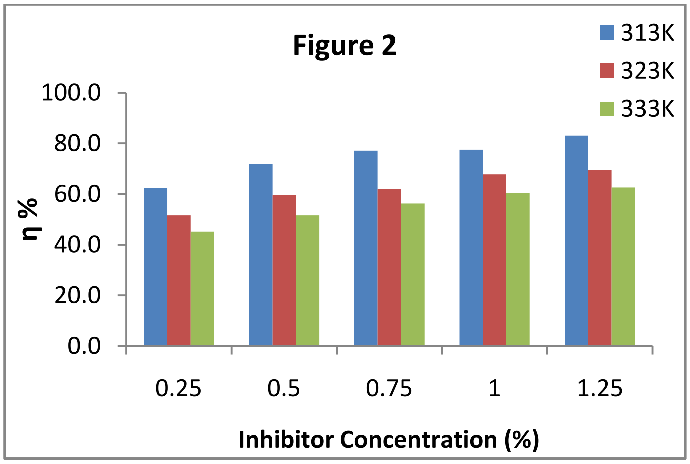

Figure 2: Relationship between inhibition efficiency η(%) and concentration of extracts in 0.5M HCl.

IJARSE
ISSN (O) 2319 - 8354
ISSN (P) 2319 - 8346

The inhibition efficiency of the Calotropis Gigantea extract is increases with the inhibitor concentration e,g, in 0.5M HCl the η% was found to be 62.4, 71.8, 77.10, 77.40 and 83.1 % with respect to 0.25, 0.50, 0.75, 1.0 and 1.25% inhibitor concentration respectively at 313K. The degree of surface coverage against inhibitor concentration (Figure 2) indicated that 1.25% inhibitor concentration has the highest protection efficiency since the highest degree of surface coverage by the inhibitor occurred at 313K. The enhanced effectiveness of the inhibitor with concentration is explicable in the light of the extent of adsorption of the inhibitor molecules on the metallic surface. It may be assumed that the film formed by the adsorbed molecules of the inhibitors on the metal surface is the sole criteria for lowering the surface area of cathodic and anodic reactions.

Table I: Thermodynamics parameter surface coverage area (θ), heat of adsorption (Q_{ads}) and entropy of adsorption (ΔS°_{ads}) and enthalpy of adsorption (ΔH°_{ads})Energy of activation (E_a) of aluminum in 0.5 M hydrochloric acid in presence of Calotropis Gigantea.

Effective area of specimen: 0.2259 sq.dm. Immersion Period : 2 h Acid Concentration : 0.5 M

Inhibitor	I. C %	θ 313 K	θ 323 K	θ 333 K	(Q_{ads}) kJ.mol^{-1} 313-323K	(Q_{ads}) kJ.mol^{-1} 323-333K	ΔH°_{ads} kJ.mol^{-1} Mean	ΔS°_{ads} kJ.mol^{-1} Mean	ΔG°_{ads} kJ.mol^{-1} Mean	Ea kJ.mol^{-1} Mean	Ea from Arrhenius plot kJ.mol^{-1}
Blank	-	-	-	-	-	-	27.51	-	-	30.15	30.60
Calotropis Gigantea (Ankado)	0.25	0.624	0.516	0.451	-37.23	-23.22	47.70	0.2575	-8.62	46.39	47.04
	0.50	0.718	0.597	0.516	-45.67	-29.18	49.43	0.2594	-7.66	53.32	54.05
	0.75	0.771	0.620	0.563	-61.04	-21.09	58.39	0.2873	-7.08	57.72	58.78
	1.00	0.774	0.677	0.603	-41.35	-28.92	51.62	0.2643	-6.70	54.46	55.14
	1.25	0.831	0.694	0.626	-64.80	-27.28	58.11	0.2847	-6.57	64.02	65.11

Several isotherms including Langmuir, Temkin and Freundlich isotherms were employed to fit the experimental data. The graph of the ratio of concentration to surface coverage (*C/*θ) against concentration (*C*) showed a straight line for tested inhibitor indicating that the adsorption of the added inhibitors followed the Langmuir adsorption isotherm. (Figures 3)

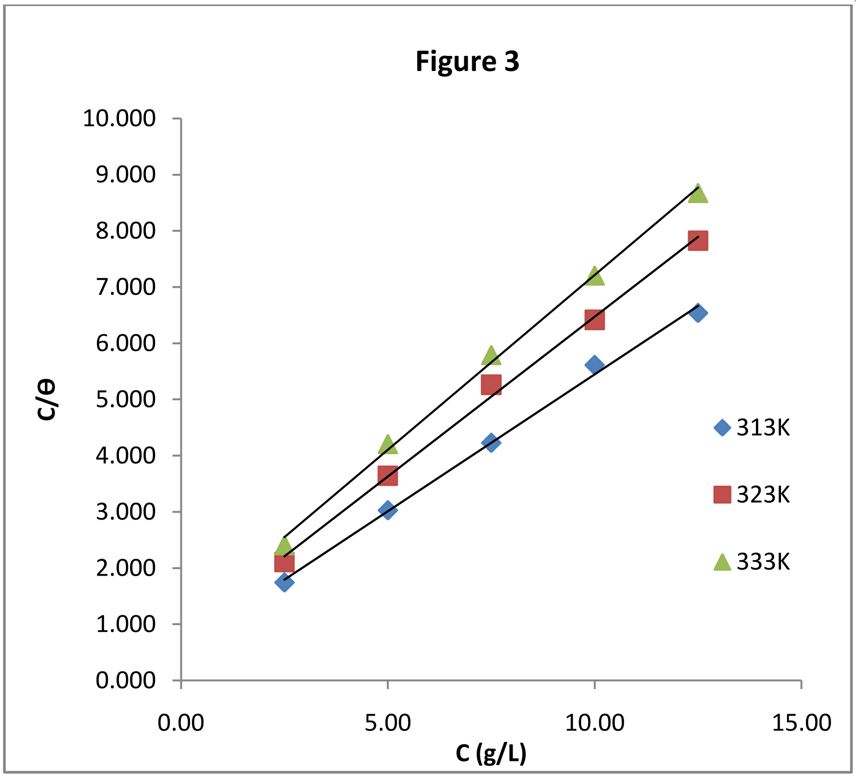

Figure 3: Plot of C/θ versus C (g/L) for Aankado leaf extract in 0.5M HCl for 2 h at different temperature.

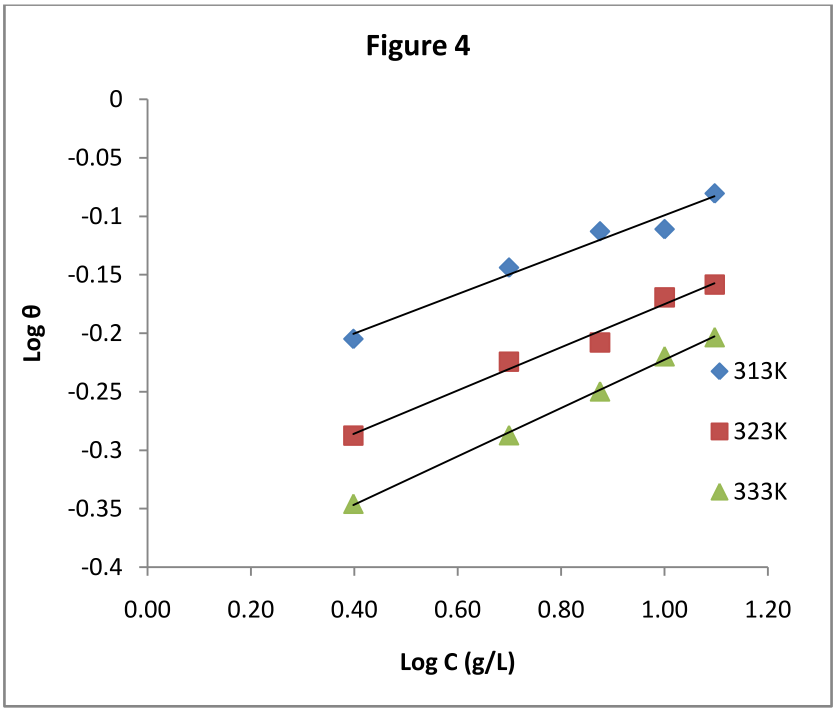

Figure 4: Plot of Log (ϴ) versus Log C (g/L) for Aankado leaf extract in 0.5M HCl for 2 h at at different temperature. (Freundlich adsorption isotherm)

IJARSE
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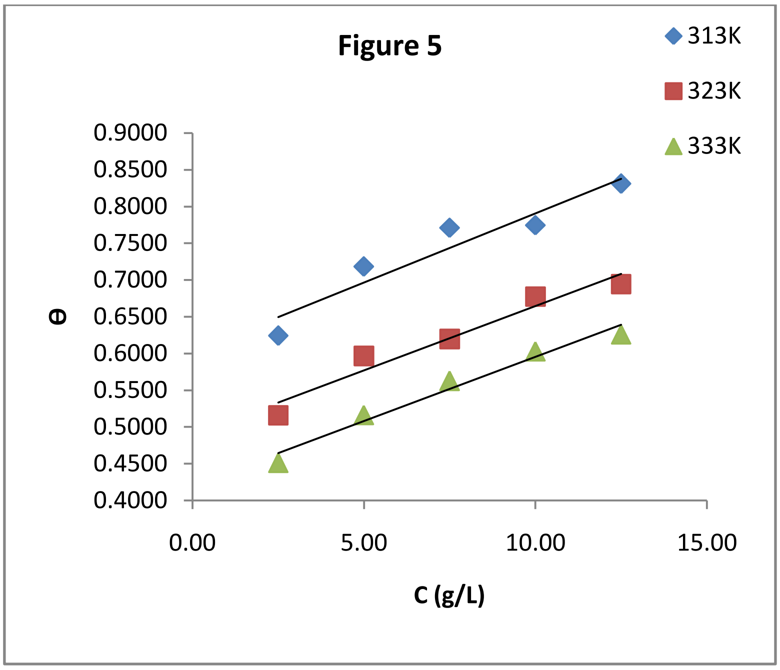

Figure 5: Plot of θ versus C (g/L) for Ankado leaf extract in 0.5M HCl for 2 h at different temperatures (Temkin's adsorption isotherm)

Table II: Some parameters of the linear regression of Freundlich adsorption isotherm for aluminum corrosion in 0.5M HCl solution containing leaf extract.

		313K	323K	333K
Calotropis	**Intercept**	-0.267	-0.359	-0.429
Gigantea	**K_{ads} ($g^{-1}L$)**	0.5408	0.4375	0.3724
(Ankado)	**ΔG^0_{ads} (kJ mol^{-1})**	-8.85	-8.857	-8.39

The degree of surface coverage (θ) and inhibitors concentration (C) can be represented by the following Freundlich adsorption isotherm [28]:

$$\log\theta = n\log C + \log K \qquad (4)$$

'K_{ads}' is the equilibrium constant for adsorption and the value of 'n' lies in range 0 to 1. Figure 4 shows the plots of Log (θ) versus Log (C) to be linear, with intercept Log K, which indicates that the experimental data fit with the Freundlich adsorption isotherm, showing that the adsorption of extract of Calotropis Gigantea on the surface of the aluminum obeys Freundlich's adsorption isotherm. The values of K_{ads} were evaluated from the intercept of the graph and presented in Table II. K_{ads} is related to the standard free energy of adsorption (ΔG^0_{ads}) and are reported in Table 2, using following equation [29, 30]:

$$K_{ads} = \frac{1}{55.5}\exp^{-\Delta G^0/RT} \qquad (5)$$

Where 55.5 is the molar concentration of H_2O in the solution, 'R' is the universal gas constant while 'T' is the absolute temperature.

IJARSE
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Plot a graph surface coverage, θ versus inhibitor concentration, C in g/l gives straight line (Figure 5) showing that the adsorption of the compound on the aluminum surface from 0.5M HCl obeys also Temkin's adsorption isotherm. It is also found that the degree of adsorption of the inhibitors increases with their concentration.

Table III: Correlation coefficient R^2 of various adsorption isotherms obtained from inhibitors for aluminum corrosion in 0.5M HCl solution containing leaf extract.

		313K	323K	333K
Calotropis Gigantea	**Temkin**	0.910	0.948	0.970
(Ankado)	**Freundlich**	0.971	0.983	0.998
	Langmuir	0.996	0.996	0.997

The linear plot with high correlation coefficient (see Table III) and slope of about unity clearly indicates that the surface adsorption process of Calotropis Gigantea extract on the aluminum alloy surface obey the all three Langmuir, Freundlich and Temkin adsorption isotherm. It revealed that physical adsorption occurred.

The results clearly showed that the inhibition mechanism involves blocking of the aluminum surface by inhibitor molecules via adsorption. Further clarification of adsorption mechanism from the experimental data the predominant adsorption mode will be dependent on factors such as the extract composition, chemical changes to the extract and the nature of the surface charge on metal. A negative surface charge will favour the adsorption of cations whereas anion adsorption is favoured by a positive surface charge. The ability of Cl^- ions in hydrochloric acid to be strongly adsorbed on the metal surface and hence facilitate physical adsorption of inhibitor cations is an important consideration [31].

The values of the free energy of adsorption (ΔG^0_{ads}) were calculated by the following equation (6) and reported in Table I. Where and C is the inhibitor concentration.

$$LogC = Log\left(\frac{\theta}{1-\theta}\right) - LogB \qquad (6)$$

Where, $LogB = -1.74 - \Delta G^0_{ads}/2.303RT$ and C is the inhibitor concentration. The mean ΔG^0ads values are negative almost in at all cases and lie in the range of -8.62 to -6.57 kJ mol^{-1}. It is evidence that the inhibitor concentration increases the values of ΔG^0ads are decreasing in order (less negative), revealed that the most efficient inhibitor shows more negative ΔG^0ads value. This suggests that they be strongly adsorbed on the metal surface. Generally, values of ΔG^0_{ads} less negative than -20 kJ. mol^{-1} indicate physical adsorption while those more negative than -40 kJ. mol^{-1} indicate chemical adsorption [32, 33]. The values ΔG^0_{ads} obtained in this experiment being less negative than -20 kJ. mol^{-1} tied with a decrease in the inhibition efficiency with increase in temperature indicates a physical adsorption process. The values of heat of adsorption (Q_{ads}) were calculated by the following equation.

$$Q_{ads} = 2.303R\left[Log\left(\frac{\theta_2}{1-\theta_2}\right) - Log\left(\frac{\theta_1}{1-\theta_1}\right)\right] X \left[\left(\frac{T_1T_2}{T_2-T_1}\right)\right] \qquad (7)$$

IJARSE
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From Table 1, it is evident that in all cases, the (Q_{ads}) values are negative and ranging from -21.09 to -64.80 kJ.mol^{-1}. The negative values show that the adsorption, and hence the inhibition efficiency, decreases with a rise in temperature.

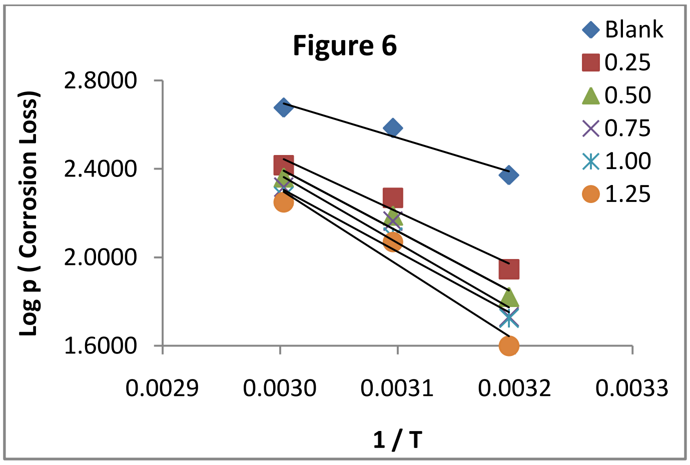

Figure 6: Arrhenius plots for aluminum in 0.5 M HCl in absence and presence of Ankado leaves ark.

Energy of activation (E_a) has been calculated with the help of following Arrhenius equation (8). Where, P_1 and P_2 are the corrosion rate at temperature T_1 and T_2 respectively.

$$Log\frac{P_2}{P_1} = \frac{Ea}{2.303R}\left[\left(\frac{1}{T_1}\right)-\left(\frac{1}{T_2}\right)\right] \quad (8)$$

Mean 'E_a' value were calculated by using eq. (8) for aluminum alloy in 0.5M HCl is 30.15 kJ. mol^{-1}, while in acid containing inhibitors, the mean E_a values are found to be higher than that of uninhibited system (Table I). The higher Ea values in the presence of inhibitor compared to the blank coupled with a decrease in the inhibition efficiency with increase in temperature can be interpreted as an indication of physical adsorption of the inhibitor on the metal surface [34]. Higher values of Ea in the presence of extract can also be correlated with the increase in thickness of the double layer that enhances the Ea of the corrosion process [35]. From the slopes of log p versus 1/T (Figure 6) energy of activation (E_a) has been calculated which are found to be quite similar that of the calculated from equation (8) and it can be shown in the Table I. Where ‘p’ is the corrosion rate and ‘T’ is the absolute temperature.

The enthalpy of adsorption (ΔH^0_{ads}) and entropy of adsorption (ΔS^0_{ads}) were calculated using the following equation (9) and (10).

$$\Delta H^0 ads = Ea - RT \quad (9)$$

$$\Delta S^0_{ads} = \frac{\Delta H^0_{ads} - \Delta G^0_{ads}}{T} \quad (10)$$

The values of ΔH^0_{ads} are positive, indicating the endothermic nature of the reaction suggesting that higher temperature favors the corrosion process [36]. The values of ΔS^0_{ads} are also positive, confirming that the corrosion process is entropically favorable [37]. In the present study general type of corrosion occurs predominately and less pitting. Calotropis Gigantea leaves extract contain significantly higher concentrations of alkaloids, fatty acids and N and O containing compounds. the major chemical constituents Calotropis Gigantea are Laurane, Saccharose, B-amyrin; A & B calotropeols; holarrhetine, Cyanidin-3-rhamnoglucoside; Taraxsterol isovalerate; Giganteol; Calotroposide; Calactin, Calotoxin; Calotropins DI and DII, Gigantin [38] (http://easyayurveda.com, 2013). The inhibitive effect is attributed due to these photochemical present in the extract. The adsorption of these organic molecules occurred due to the formation of a links between aluminum atoms, involving the displacement of water molecules from metal surface, and the lone pairs present on N- and O- atoms of the heterocyclic rings. It can be concluded that, due to the suitable inhibitive characteristics of compounds present in Calotropis Gigantea leaves extract is effective inhibitor for aluminum corrosion. Alka Sharma et al. [39] also support the above statement.

II. CONCLUSION

As a constant inhibitor concentration, the inhibition efficiency of Calotropis Gigantea leaves extract decreases as the concentration of acid increases. As the inhibitor concentration increases inhibition efficiency increases and corrosion rate decreases. 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. In all cases, the value of heat of adsorption (Q_{ads}) and the value of free energy of adsorption (ΔG^0_a) is negative. The Value of change of enthalpy (ΔH^0_a) and entropy of adsorption (ΔS^0_a) is positive. A mean value of 'E_a' in inhibiting acid is higher than the value of 'Ea' in acid only.

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