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The Effect of *Acanthus montanus* Leaves Extract on Corrosion of Aluminium in Hydrochloric Acid Medium

G. I. Udom[1], G. A. Cookey[1*] and A. A. Abia[2]

[1]Department of Chemistry, Rivers State University, Port Harcourt, Rivers State, Nigeria.
[2]Department of Pure and Industrial Chemistry, University of Port Harcourt, Port Harcourt, Rivers State, Nigeria.

Authors' contributions

This work was carried out in collaboration among all authors. Author GIU performed the experiment and statistical analyses, wrote the protocol and the first draft of the manuscript. Author GAC supervised the experiments and statistical analyses of the study and edited the manuscript. Author AAA designed the study and edited the manuscript. All authors read and approved the final manuscript.

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ABSTRACT

The inhibitive effect of *Acanthus montanus* leaves extract on aluminium corrosion in hydrochloric acid solutions was studied using weight loss technique. It was found that the leaves extract retards the acid induced corrosion of aluminium. Inhibition efficiency was observed to increase with increase in inhibitor concentration but decreased with rise in temperature. Thermodynamic studies revealed that the corrosion inhibition may be due to the spontaneous physical adsorption of the extract constituents on aluminium surface. Adsorption was found to obey Temkin isotherm. A first–order mechanism has been deduced from the results at all temperatures studied.

Keywords: Aluminium corrosion; Acanthus montanus; weight loss; adsorption; thermodynamics; kinetics.

**Corresponding author: E-mail: cookey.grace@ust.edu.ng;*

1. INTRODUCTION

Aluminium is one of the most essential and widely used metals in many industries such as automotive, aerospace, construction and power generation. It is a hard, strong and highly electropositive metal. The most important feature of aluminium is its resistance to corrosion due to the presence of a thin, adherent and positive surface [1]. However, when exposed to the action of acid in industrial process where acid solutions are normally used for pickling, industrial acid cleaning, oil well oxidizing, acid descaling, chemical and electrochemical etching of aluminium, the oxide on the metal surface dissolves and makes the metal surface susceptible to attack in aggressive media which usually leads to severe loss of the metal due to corrosion [2]. Corrosion inhibitors are usually added in small quantities to reduce or prevent metal surface dissolution and protect the metal in acidic or aggressive environment and they are an effective method of preventing corrosion [3]. Some of the compounds used as corrosion inhibitors are toxic, non-biodegradable and expensive. This led to the use of plant parts extracts which are low cost, environmental friendly, readily available and renewable source to prevent corrosion in this study. The use of various plant parts extract as corrosion inhibitor for metals in different media has been reported by [4,5,6,7]. The potency of African Black Velvet Tamarind as inhibitor of aluminium in HCl acid solution has been studied by [8].

[1] investigated inhibition of aluminium in acidic medium by different extracts of ocimum gratissium. [2] studied the corrosion control of aluminium by *Lawsonia inermis* seed extract in acid medium. The corrosion inhibitive effects of Anigeria rebusta extract for aluminium in HCl solution was carried out by [9]. The corrosion behaviour of aluminium in HCl by date palm leaves extracts was investigated by [10]. The inhibitory effects of ethanol extract of *Vernonia amydgalina* on the corrosion of aluminium in HCl acid was carried out by [11]. *Gossipium hirsutum* L. extract as green corrosion inhibitor for aluminium in HCl solution has been reported [12]. The inhibition efficiency of garlic extract on aluminium in hydrochloric acid solutions has been carried out by [13]. Comparative study of leaves extracts for corrosion inhibition effect on aluminium alloy in alkaline medium has been exported [14]. The inhibition potentials of plants extracts are due to the presence of the chemical compounds in the extracts, including tannin, alkaloids, nitrogenous bases, carbohydrates and proteins which get adsorbed onto the metal surface, blocking the corrosion site and retarding the wearing away of the metal [15].

This study was to assess the inhibitive and adsorption properties of *Acanthus montanus* leaves extract on corrosion of aluminium in HCl acid solution. *Acanthus montanus,* which is of the family *Acanthaceae* (leopard tongue or false thistle), is a prickly herb found in the Mediterranean, Australia and USA. It is also found in the high forest Upper Guinea, West Cameroons as well as in Nigeria. *Acanthus montanus* is used in South-Western Nigeria for treatment of various ailments, like stomach upset, urinary disorders, etc. The fresh leaves are used in South-eastern Nigeria to treat boil in the finger. In Congo, the decoction of the leafy twig is used as purgative [16]. The plant parts are biodegradable and renewable materials.

Presently, and to the best of our knowledge, there is no reported work in the open literature on the effects of *Acanthus montanus* leaves extract on corrosion of aluminium and other metals in acidic solutions. Therefore, this paper reports the inhibitive effects of *Acanthus montanus* leaves on corrosion of aluminium in 2.0M HCl solution using weight loss technique.

2. MATERIALS PREPARATION

Aluminium sheet of 0.1 cm thickness and 98.95% purity was obtained and mechanically press-cut into coupons of 2.0 cm x 4.0 cm dimension in engineering workshop of the University of Port Harcourt, Rivers State, Nigeria. The coupons were polished with emery papers to obtain a clean shiny surface washed in deionized water, degreased in ethanol, dried in acetone and then weighed and stored in moisture free desiccator prior to use. The corrosive medium was 2.0M HCl of analytical grade by Sigma-Aldrich. Deionized water was used for the preparation of all reagents.

2.1 Preparation of *Acanthus montanus* Leaves Extracts

The fresh leaves of *Acanthus montanus* plant were obtained from Oruk-anam, Akwa Ibom State, South-South Nigeria. The leaves were washed thoroughly and rinsed with deionized water to remove soil impurities, sun-dried and milled to powder. From the finely powdered leaves, 50 g were placed in 500ml round bottom flask containing 99.8% methanol in a soxhlet extractor. The resulting extract was evaporated in an oven at 40℃ until drying and then weighed and stored in sample container for use. The extract was then used for phytochemical analysis and also to prepare inhibitor concentrations in dilute HCl acid solution. Five different concentrations ($5.0x10^{-2}$, $4.0x10^{-3}$, $3.0x10^{-4}$, 2.0 $x10^{-4}$ and $1.0.x10^{-6}$ M) of the inhibitor were prepared by dissolving 0.5 g of *Acanthus montanus* leaves extracts with 250 ml of 2.0M HCl acid solution in a round bottom flask and used throughout the present investigation.

2.2 Weight Loss Determination

Specimens of previously weighed aluminium coupons of 2.0 cm x 4.0 cm x 0.1 cm with a hole in them were suspended in 50 ml beakers of the test solutions in the presence and absence of the extract (inhibitor) at 303-333 K. Thermostated water bath was set to appropriate temperatures for those at 313 – 333 K. The weight loss was monitored by retrieving the coupons from the test solutions at 2 hours intervals progressively for 18 hours. Weight loss was calculated by finding the difference in the weight of coupons before and after immersion in the test solutions as shown in Equation 1. The procedure for weight loss determination was similar to that reported by [17].

$$\Delta W_B = W_i - W_f \tag{1}$$

where W_i is the weight before immersion, while W_f is the weight after immersion in the test solution. From the weight loss, the percentage inhibition efficiency (%IE), corrosion rate (CR), surface coverage (Ѳ) and rate constant (k) were calculated. (%IE) was calculated using Equation 2 [18].

$$IE\% = 100\left(\frac{\Delta W_B - \Delta W_{inh}}{\Delta W_B}\right) \tag{2}$$

Where, W_B and W_{inh} are the weight losses of aluminium coupon in the absence and presence of *Acanthus montanus* leaves extract in HCl at a particular temperature.

Corrosion rates ($gcm^{-2}h^{-1}$) of aluminium in different corrosion media were determined using the formula in Equation 3 [19].

$$CR = \frac{\Delta W}{At} \tag{3}$$

where ΔW = weight loss (g), *A* is surface area (cm^2) of adsorbent and t is the time of immersion (hours).

The degree of the adsorbent surface covered by the adsorbate (*Ѳ)* was calculated using Equation 4.

$$\theta = \left[1 - \frac{\Delta W_{inh}}{\Delta W_B}\right] \quad .... \quad (4)$$

3. RESULTS AND DISCUSSION

3.1 Effect of *Acanthus montanus* Leaves Extract on Corrosion of Aluminium in HCl (Weight Loss)

The variations of weight losses with time for aluminium in 2.0 M HCl containing different concentrations of *Acanthus montanus* leaves extract for temperatures ranging from 303 to 333K are presented in Fig. 1.

From the plots, weight losses for systems with *Acanthus montanus* leaves extract were found to be lower compared to the systems without the extracts. The figures also reveal that the tendency of weight losses decreased with increasing concentration of the extract. These results may suggest that *Acanthus montanus* leaves extract significantly inhibit the corrosion of aluminium in 2.0M HCl medium and the degree of inhibition increases with increasing concentration of the extracts. Similar results on the adsorption of Gossipium hirsutum Leaves extract as inhibitor have been obtained [20].

3.2 Effect of Extract Concentration on Inhibition Efficiency

The inhibition efficiency (IE%) of aluminium in different concentrations of *Acanthus montanus* leaves extract in 2.0M HCl between the temperature range of 303 to 333K is shown in Fig. 2.

The plot shows that inhibition efficiency increases as concentration increases at all temperatures but decreases slightly with increase in temperature. Maximum inhibition efficiency of 73.44% was observed at the highest concentration of $5.0x10^{-2}$M at 303K. Hence, increase in the concentrations of the inhibitor increased the adsorption of the inhibitor molecules on the metal surface thus retarding weight loss of aluminium coupons.

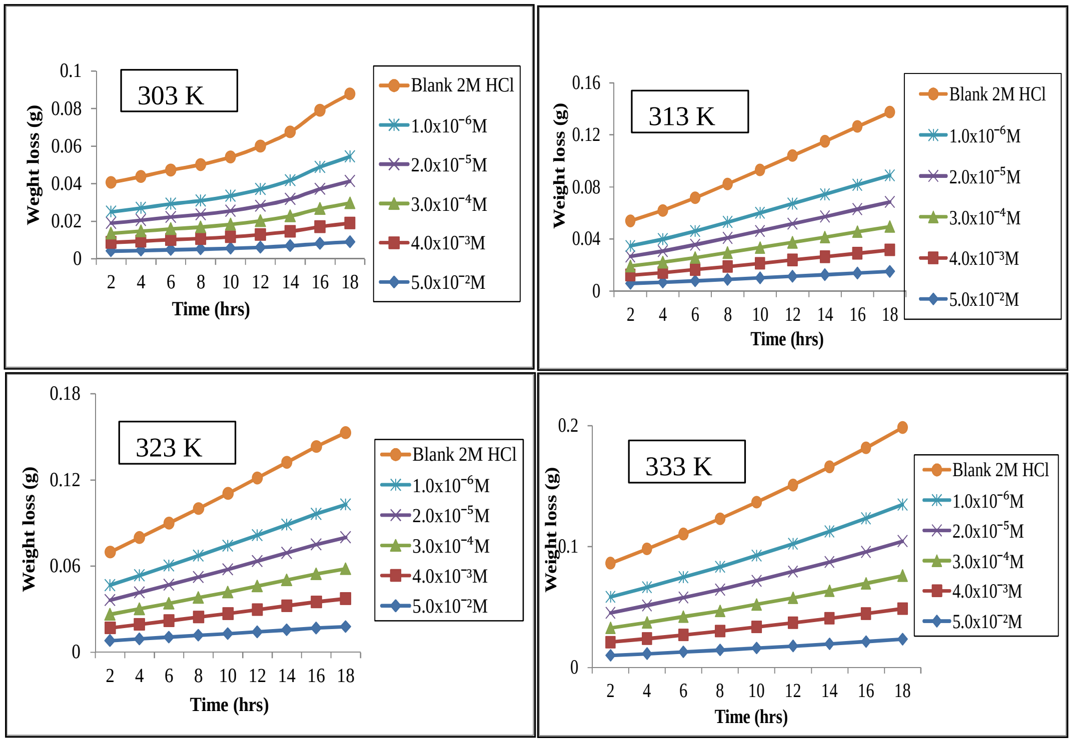

Fig. 1. Variation of weight loss (g) with time (hrs) for Aluminium Coupons in 2M HCl at different concentrations of *Acanthus montanus* extract and temperatures

The same trend was also observed by [3] in the study of corrosion inhibitive properties and adsorption behaviour of Piper guinensis extract obtained from ethanol. [21] reported the use of flavonoid (catechin) extracted from peanut skin in corrosion inhibition of mild steel in hydrochloric acid solution.

3.3 The Effect of Temperature on Corrosion Rate and Inhibition Efficiency

The effect of temperature on the corrosion rate of aluminium in the absence and presence of different concentrations of *Acanthus montanus* leaves extracts (inhibitor) is shown in Table 1. The table indicates that corrosion rate increased with increase in temperature in both cases but decreased with increasing concentrations of the inhibitor. However, at every temperature, corrosion rate is higher for uninhibited aluminium than the inhibited one. The decrease in corrosion rate for inhibited acid solutions compared to the uninhibited suggests retarding effect of *Acanthus montanus* leaves extract on the corrosion of the aluminium. Table 1 also shows the dependence of inhibition efficiency on the concentration of inhibitor extract and temperature. While % IE generally decreased with temperature rise, it increased with increasing concentration of the inhibitor. The decrease in inhibition efficiency with temperature could be attributed to desorption of the molecules *of Acanthus montanus* leaves extract from aluminium surface. Similar observations have also been reported by [3,22]. The table also shows a good correlation between % inhibition efficiency and surface coverage and both decreased with increase in temperature.

3.4 Adsorption/Thermodynamic Studies

The study of adsorption isotherms is very useful in determining the extent of interaction between inhibitor and metal surface in acidic medium. The values of θ for different concentrations of *Acanthus montanus* extract were obtained using Equation 4. Temkin adsorption isotherm (equation 5) was used to describe the adsorption of the leaves extract on aluminium metal surface.

$$\exp(-2a\theta) = K_{ad}C \tag{5}$$

where K_{ad} is the equilibrium constant of adsorption, C is the inhibitor concentration, a is the molecular interaction parameter and θ is the degree of surface coverage. The plots of θ versus C at the different temperatures are illustrated in Fig. 3. The linear relationship between Ө and log C and the good correlation coefficient (Table 2) suggest that the experimental data of *Acanthus montanus* leave extract obey Temkin adsorption isotherm [23].

The adsorption data was also applied to Langmuir and Freundlich isotherms and the plots are shown in the Supplementary information. The results show that Freundlich and Temkin isotherms describe the adsorption process better than that of Langmuir, the two previous isotherms being influenced by temperature changes while temperature variation had no effect on the Langmuir isotherm.

There is a good correlation between % inhibition efficiency and surface coverage as both decrease with increase in temperature. The plots confirm that the mechanism of corrosion inhibition is due to the formation of protective film on the metal surface and that the inhibitor was uniformly adsorbed on the metal surface. [24] had also made such observation in the inhibitive effect of water extract of the stem bark of *Anthocleista djalonensis* on acid corrosion of aluminium.

The values of Temkin parameters (Equilibrium constant, interaction parameter and correlation coefficient) extrapolated from the curves at the different temperatures are tabulated in Table 2.

Interaction parameter values are negative at all temperatures which is indicative of repulsive interactions between the molecules of *Acanthus montanus* leave extract and Aluminium surface. Values of equilibrium constant decrease with temperature rise which is in agreement with the decreasing value of IE (%) as temperature increases.

3.5 Activation Energy (E_a)

The activation energy parameters for the corrosion process were obtained using Arrhenius-type equation (Equation 6).

$$LogCR = \log A - \frac{E_a}{2.303RT} \quad (6)$$

Where, CR is the corrosion rate, E_a is the apparent activation energy, T is the absolute temperature, R is the universal gas constant and A is the Arrhenius pre-exponential factor. The values of Ea were determined by plotting log CR vs. I/T (Fig. 4).

Table 1. Corrosion rate (CR ($gcm^{-2}day^{-1}$) x10^{-3} of mild steel in *Acanthus montanus* extract and 2.0M HCl at different temperatures

T (K)	CR			
	303	313	323	333
Conc.(M)				
Blank	18.78	27.81	30.98	37.25
1.0 x 10^{-6}	7.27	10.93	13.75	17.67
2.0 x 10^{-5}	6.54	10.77	13.25	16.58
3.0 x 10^{-4}	5.96	10.17	12.58	15.92
4.0 x 10^{-3}	5.59	9.39	11.75	14.72
5.0 x 10^{-2}	5.00	8.50	10.75	13.67

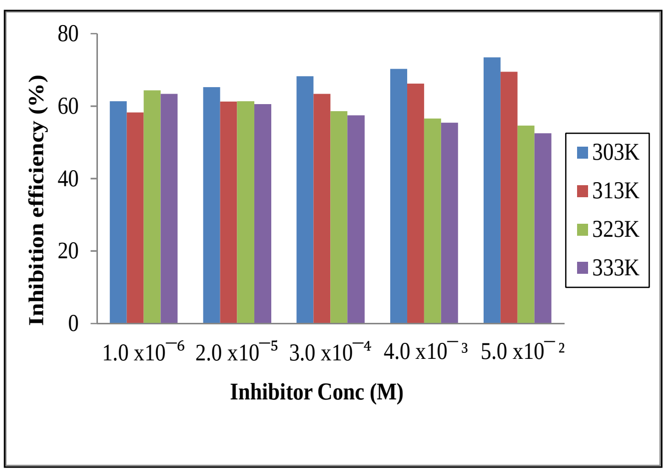

Fig. 2. Varaition of percentage inhibition efficiency wih inhibitor concentrations (M) for the corrosion of Aluminium coupons

Table 2. Surface coverage and Inhibition efficiency (IE %) of *Acanthus montanus* extract in 2.0M HCl at different temperatures

	IE (%)				Surface coverage (θ)			
T (K)	**303**	**313**	**323**	**333**	**303**	**313**	**323**	**333**
Conc.(M)								
1.0×10^{-6}	61.34	58.30	54.43	52.55	0.61	0.58	0.54	0.52
2.0×10^{-5}	65.23	61.32	56.54	55.46	0.65	0.61	0.56	0.55
3.0×10^{-4}	68.30	63.37	58.63	57.36	0.68	0.63	0.58	0.57
4.0×10^{-3}	70.25	66.25	61.39	60.47	0.70	0.66	0.61	0.60
5.0×10^{-2}	73.44	69.45	64.43	63.39	0.73	0.69	0.64	0.63

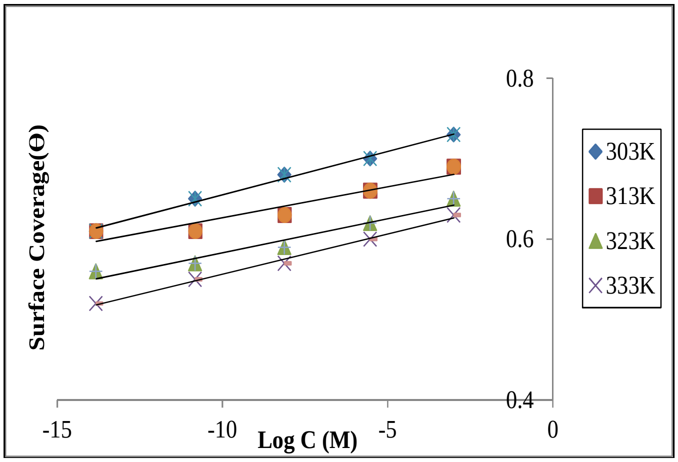

Fig. 3. Temkin Isotherm plot of aluminum surface coverage versus log inhibitor concentration (M)

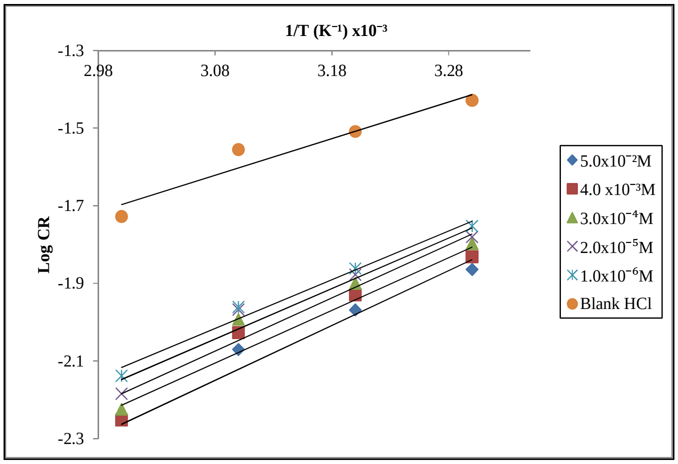

Fig. 4. Plot of log corrosion rate (CR) versus inverse of absolute temperature (1/T)

The slope of the straight line obtained was –Ea/2.303R from where Ea values were evaluated. Table 4 shows the calculated values of E_a.

The data of Table 4 shows that the E_a value for the blank is 18.090 $kJmol^{-1}$ which increased to 27.037 kJ/mol^{-1} in the presence of *Acanthus montanus* leaves extracts at the maximum concentration. This observation may indicate that the adsorbed organic extract provided a physical barrier to the corrosion of aluminium coupons, the strength of which increased with increasing concentration of the extract. [25,26] reported that values of E_a > 80 $kJmol^{-1}$ indicate chemical adsorption while those < 80$KJmol^{-1}$ result from physical adsorption. Thus, the E_a values in this study support the fact that the *Acanthus montanus* leaves extracts were physically adsorbed on the surface of aluminium coupons.

Table 3. Temkin parameters for the adsorption of *Acanthus montanus* leave extract on Aluminium surface at 303-333 K

T (K)	a	K_{ads}	R^2
303	-0.02	1.37 $\times 10^{33}$	0.992
313	-0.02	1.25 $\times 10^{31}$	0.993
323	-0.018	8.81 $\times 10^{31}$	0.981
333	-0.02	3.09 10^{28}	0.993

Table 4. The Activation Energy (E_a) values at the various concentrations of the *Acanthus montanus* leaves extract

Extract concentration	E_a (kJ/mol)
Blank	18.090
1.0×10^{-6}	24.064
3.0×10^{-4}	25.688
4.0×10^{-3}	26.000
5.0×10^{-2}	27.037

3.6 Determination of Enthalpy and Entropy of Adsorption

Thermodynamic parameters such as enthalpy (ΔH) and entropy (ΔS) of adsorption of *Acanthus montanus* leaves extract on aluminum surface may be evaluated from temperature effects (equation 7) in the absence and presence of the extract [27].

$$Log\left(\frac{CR}{T}\right)=\left[\left(\log\frac{R}{Nh}\right)+\frac{\Delta S}{2.303R}\right]-\frac{\Delta H}{2.303RT} \quad (7)$$

Here CR is corrosion rate, T is absolute temperature, R is the universal gas constant while N and h are Avogadro's and Planck's constant respectively. A plot of log CR/T vs. 1/T gives a straight line with slope,-ΔH/(2.303R) and intercept, [(log R/Nh) + ΔS/(2.303R)].

The derived values of enthalpy and entropy of corrosion are shown in Table 5. The results presented in this table show that the ΔH values are positive in the absence and presence of *Acanthus montanus* leaves extract which reflect the endothermic nature of the aluminium dissolution process while the values of ΔS in both solutions are large and negative, indicating spontaneous and feasible reaction forming activated complex that represents association rather than dissociation steps. Similar observations have been reported in literature [13].

Table 5. Enthalpy (ΔH) and Entropy (ΔS) values of corrosion at the various concentrations of the extract

Extract Conc.(M)	ΔH (kJ/mol)	−ΔS (J/mol)
Blank	15.212	189.185
1.0×10^{-6}	17.414	202.228
2.0×10^{-5}	22.209	218.792
3.0×10^{-4}	23.649	223.838
4.0×10^{-3}	23.365	223.543
5.0×10^{-2}	24.426	227.669

3.7 Gibb's Free Energy of Adsorption of *Acanthus montanus* Leaves Extract on Aluminium

The equilibrium constant of adsorption (K_{ads}) obtained from the intercepts of θ vs log C Temkin isotherm plot is related to the free energy of adsorption (ΔG_{ads}) as shown in Equation 8.

$$\Delta G_{ads} = 2.303RT\log(55.5K_{ads}) \quad (8)$$

In equation 8, R is the universal gas constant, T is the absolute temperature and 55.5 is molar concentration of one liter of water. The calculated values of the free energy of adsorption are presented in Table 6.

Table 6. Calculated values of the free energy of adsorption at the various temperatures

Temperature (K)	$-\Delta G_{ads}$ (kJ/mol)
303	9.4381
313	9.5387
323	9.7035
333	9.9563

It is observed that the values of ΔG_{ads} are negative at all temperatures, showing that the *Acanthus montanus* leaves extract was strongly adsorbed onto the aluminium surface. [28] have also reported similar results.

Literature review reveals that negative values of 20 kJ/mol or less of ΔG_{ads} are consistent with electrostatic interaction between molecules of charged adsorbent and adsorbate that arise from physisorption processes. Conversely, [29,30] have attributed values around or less than -40kJ/mol to charge transfer from the inhibitor molecules to the metal surface or sharing of electrons between them. Subsequently, co-ordinate type bonds are formed (chemisorption). The results of free energy of adsorption obtained from this study are thus consistent with previous works from literature and indicate that the adsorption of *Acanthus montanus* leaves extract on aluminium surface was through physisorption mechanism.

3.8 Kinetic Study

The corrosion of aluminium in HCl is a heterogeneous one that comprises of anodic and cathodic reactions at the same or different rates. On this basis, kinetic analysis of the data was considered necessary. In this study, log (W_i–ΔW) was plotted against time (hrs) as shown in Fig. 6 (where W_i and ΔW are as defined earlier and k, is the rate constant. The plot of log (W_i–ΔW) against t gives a linear graph which confirms a first order reaction mechanism.

The rate constant (k) was calculated from first order rate Equation (9).

$$k = \left(\frac{2.303}{t}\right)\log\frac{W_i}{W_f} \tag{9}$$

The results obtained (Table 7) show that the rate constant of corrosion decreased with increase in concentration of the inhibitor. The half-life ($t_{½}$) of the corrosion reaction was calculated from first order half-life equation (Equation 10).

$$t_{1/2} = \frac{0.693}{k} \tag{10}$$

The $t_{1/2}$ values were observed to increase with increase in concentration of the inhibitor and decreased as temperature increased from 303 to 333K (Table 6). The increase in half-life with concentration of the leaves extract indicates better protection of aluminium coupons by the inhibitor [31].

3.9 Phytochemical Constituents of *Acanthus montanus* Leaves Extract

The chemical constituents present in *Acanthus montanus* leaves are listed in Table 7. As reported earlier [32], these compounds are easily hydrolysable and can be adsorbed on the aluminium surface through the lone pair of electrons present on the constituent oxygen atoms. However, they contain multifunctional groups which create charge and mass transfer barrier leading to decrease in the interaction of the metal with the corrosive media. Consequently, film layers are formed which essentially block the discharge of H^+ subsequent dissolution of the metal ions [19]. As a result, the corrosion rate of aluminum decreased in the presence of the inhibitor extract.

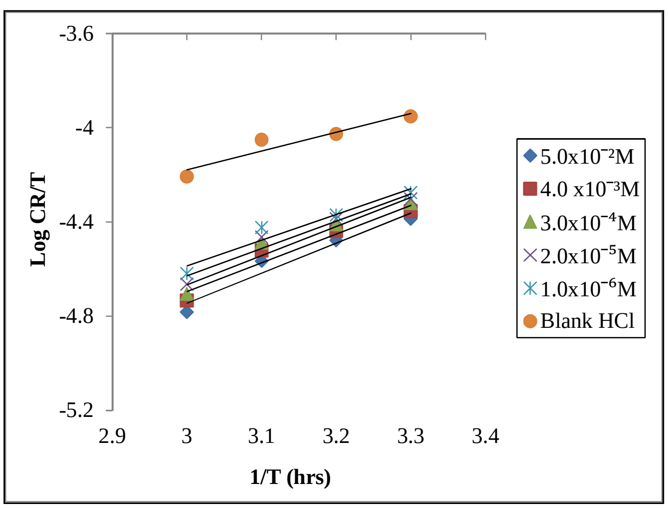

Fig. 5. Plots of Log CR/T vs. 1/T for the corrosion of aluminum in 2M HCl at different concentrations of the extract

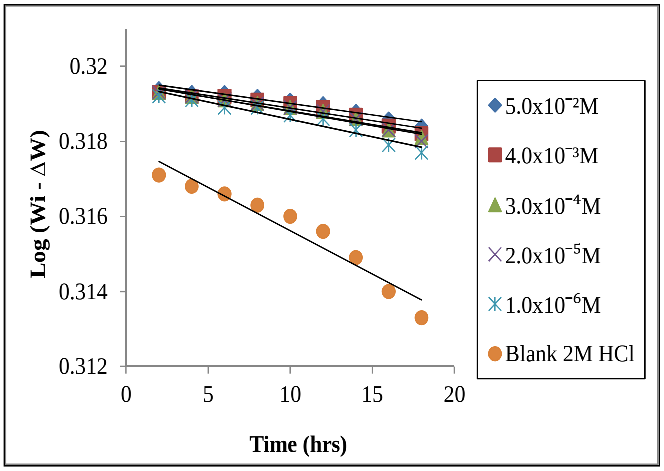

Fig. 6. Variation of Log (Wi-ΔW) with time (hrs) for the corrosion of Aluminium coupons in 2.0M HCl solution with and without *Acanthus montanus* extract at 303 K

Table 7. Rate constant (k) and half-life parameters at various concentrations of the inhibitor extract

Temp.(K)	k (day^{-1}) x 10^{-3}				Half life (days)			
	303	313	323	333	303	313	323	333
Extract Conc.(M)								
Blank	9.06	13.59	14.69	17.17	76.51	50.99	47.16	40.35
1.0 x 10^{-6}	3.59	5.64	6.57	7.69	192.78	122.93	105.49	90.12
2.0 x 10^{-5}	3.15	5.23	6.30	7.75	219.71	132.39	109.97	89.44
3.0 x 10^{-4}	2.87	4.97	5.95	7.36	241.03	139.39	116.38	94.19
4.0 x 10^{-3}	2.72	4.58	5.56	6.78	255.21	151.33	124.71	102.25
5.0 x 10^{-2}	2.39	4.13	2.66	6.28	289.28	162.83	260.06	110.43

Table 8. The phytochemical Results for Methanol extract of *Acanthus montanus* leaves

Phytochemical constituents	Results
Alkaloid	+++
Tannin	+++
Saponin	+++
Flavonoid	++
Terpenoids	++
Cardiac glyosides	+++
carbohydrate	+++
protein	+++
steroids	-
Anthraquinone	-
Fixed oils	+++

- =Absent, ++ = Moderately present, +++ = Present in concentration

4. CONCLUSION

The inhibitive effect of methanol extract of *Acanthus montanus* leaves on aluminum metal in HCl medium has been studied at different temperatures and inhibitor concentrations. Results obtained show that inhibition efficiency increased with increase in concentration of the extract but decreased with increase in temperature.

The thermodynamic study of the inhibition process yielded negative values of free energy of adsorption indicating spontaneity of *Acanthus montanus* extract adsorption on aluminum surface. The mechanism of physical adsorption has been proposed and a first order reaction obtained from kinetic treatment of the experimental data.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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