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Aluminium Corrosion Inhibition by *Maesobatrya barteri* Root Extract in Hydrochloric Acid Solution

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Authors' contributions

This work was carried out in collaboration between both authors. Author OUA designed the study, performed the statistical analysis, wrote the protocol, and wrote the first draft of the manuscript. Author IEM managed the analyses of the study and the literature searches. Both authors read and approved the final manuscript.

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ABSTRACT

Maesobatrya barteri root extract inhibited the corrosion of aluminium in hydrochloric acid solutions by both gravimetric and thermometric methods. The inhibition efficiency increased with increase in root extract concentration but decreased with increase in temperature. The adsorption of the root extract on aluminium surface obeyed the Langmuir adsorption isotherm. The calculated thermodynamic parameters revealed that the corrosion inhibition process was both endothermic and spontaneous. Physical adsorption has been proposed for the adsorption of *Maesobatrya barteri* root extract on aluminium surface.

Keywords: Corrosion inhibition; Langmuir isotherm; aluminium; Maesobatrya barteri; physisorption.

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1. INTRODUCTION

The corrosion of metals due to exposure to aqueous and non - aqueous solvents is a worldwide phenomenon [1]. The destructive effects of corrosion in the building, construction, petroleum and aviation industries, amongst others, are very glaring. The use of corrosion inhibitors is the most cost effective means of combating corrosion. Several synthesized organic compounds containing N, S, and/or O atoms have been reported as promising inhibitors for aluminium corrosion in acid media [2–5]. These compounds inhibit the corrosion process by adsorbing on the metal surface, forming protective thin films which reduce/stop the electron transfer process on the metal surface. However, many of these compounds are toxic [6], expensive, environmental unfriendly [7] and not readily available.

Research efforts in recent times have shifted to the use of plant extracts as inhibitors of metal corrosion in acidic and alkaline media. Plants are rich sources of organic nitrogen, sulphur and oxygen. Some of the phytochemicals extracted from plants include tannins, alkaloids, terpenes, saponins, cardiac glycoside, deoxy sugars, etc. Inhibitors extracted from plants are cheap, non – toxic, readily available and environmentally friendly. Several researchers have reported the inhibition of aluminium corrosion in acidic medium using plant extracts [8–12], though with varying inhibition efficiencies.

Maesobotrya barteri (English name: Bush cherry; Efik/Ibibio name: Nyanyatet) is a medicinal plant with edible fruits used traditionally by the people of south eastern Nigeria for the treatment of various ailments. The sap obtained from the crushed roots of the plant is used for the treatment of some skin diseases [13]. There is no reported work on the inhibition of aluminium corrosion by *Maesobotrya barteri* extracts. This work aims at inhibiting the corrosion of aluminium in hydrochloric acid solutions by *Maesobotrya barteri* root extract.

2. MATERIALS AND METHODS

2.1 Test Materials

The aluminium sheet used for this work was obtained from System Metals Industries Limited, Calabar, Nigeria. The aluminium sheet used for weight loss tests was mechanically press – cut into coupons of 4.0 cm x 5.0 cm containing a small hole near the upper edge for the insertion of glass hooks. Each coupon had a total surface area of 40.0 cm^2. The aluminium coupons used for the thermometric tests were 2.0 cm x 5.0 cm in dimension.

The aluminium coupons were used as cut without further polishing. However, they were degreased in absolute ethanol, dipped in acetone and dried in air and then stored in moisture – free desiccators before use in corrosion studies.

Analar grade reagents were used throughout for the work. Hydrochloric acid and ethanol were of Sigma – Aldrich Laboratories, Germany. Acetone was of BDH Chemicals, England. Deionised water was used throughout for the preparation of solutions.

2.2 Preparation of *Maesobatrya barteri* Root Extract

Fresh roots of *Maesobatrya barteri* were collected from the Main Campus of University of Uyo, Uyo, Nigeria. They were washed, cut into small pieces and air – dried at 30℃ for seven days. They were then ground to powder. The dried ground samples of *Maesobatrya barteri* root were macerated with 90% ethanol for seven days at room temperature in a large glass trough with cover. The mixture was then filtered. The filtrate was evaporated at 40℃ in a water bath to constant weight, leaving a brown coloured extract in the beaker. Extract concentrations of 0.5 g/L, 1.0 g/L, 1.5 g/L, 2.0 g/L, and 4.0 g/L respectively in 0.5M HCl solution were used for the gravimetric studies at 30℃ – 60℃. The same extract concentrations were used in 2 M HCl solution for the thermometric tests.

2.3 Gravimetric Methods

Previously weighed aluminium coupons were suspended by glass hooks and immersed in 0.5 M HCl solution in separate 100 ml beakers in thermostatic water bath maintained at 30℃, 40℃, 50℃ and 60℃, respectively. The coupons were retrieved from their corroding solution after 4 hours, dipped into concentrated nitric acid and washed several times in water. The coupons were then dipped in acetone and dried in air before reweighing [14]. The difference in weight was taken as weight loss.

The second stage of the weight loss method involved the use of inhibitor (*Maesobatrya barteri* root extract). One aluminium coupon per beaker was suspended by a glass hook and immersed in 100 ml of 0.5M HCl solution containing different concentrations of the extract (0.5 g/L, 1.0 g/L, 1.5 g/L, 2.0 g/L, and 4.0 g/L). Each coupon was retrieved from HCl – extract medium after 4 hours, washed and reweighed. The weight loss was recorded. The weight loss was used to calculate the inhibition efficiency (%I) using the formula [15]:

$$\%I = \left(\frac{W_0 - W_1}{W_0}\right) \times 100 \quad (1)$$

where W_1 and W_0 are the weight losses of aluminium in HCl solution, with and without inhibitors, respectively.

The corrosion rate, CR (mg cm^{-2} hr^{-1}), of aluminium was obtained using the equation:

$$CR = \frac{W}{A\,t} \quad (2)$$

where W is the weight loss of aluminium coupon (mg), A is the surface area (cm^2) while t is the immersion time (hr).

2.4 Thermometric Methods

The reaction vessel and procedure for determining the corrosion behaviour by this method has been described by other researchers [3,16]. One aluminium coupon (2 cm x 5 cm) was used for each test. The initial temperature in all experiment was kept at 30.0℃. The variation of temperature with time was monitored per minute in a calibrated thermometer (0℃ - 110℃) to the nearest ±0.1℃. The temperature kept rising to a maximum value before decreasing. This technique allowed for evaluation of reaction number (RN) defined as:

$$RN\ (°C/min) = \frac{T_m - T_i}{t} \quad (3)$$

where T_m and T_i are the maximum and initial temperatures. The inhibition efficiency (%I) was calculated from percentage reduction in reaction number [17]:

$$\%I = \left(\frac{RN_0 - RN_1}{RN_0}\right) \times 100 \quad (4)$$

where RN_0 is the reaction number in the absence of inhibitor and RN_1 is the reaction number in the presence of inhibitor.

3. RESULTS AND DISCUSSION

3.1 Effect of Extract Concentration on Inhibition Efficiency

Fig. 1 shows the variation of inhibition efficiency with concentration of *Maesobatrya barteri* root extract in 0.5 M HCl solution by gravimetric measurements. It is observed that, at a particular temperature, the inhibition efficiency increased with increase in the concentration of the root extract. The thermometric results for aluminium corrosion in 2 M HCl solution containing *Maesobatrya barteri* root extract are illustrated in Fig. 2. Inspection of Fig. 2 shows that as the concentration of the root extract increases, the time required to reach the maximum temperature (t) increases while the maximum temperature (T_m) decreases. Table 1 shows the calculated values of reaction number (RN) and inhibition efficiency (%I) for aluminium corrosion in 2 M HCl containing *Maesobatrya barteri* root extract. It is observed that the more effective the extract (inhibitor) concentration, the lesser the reaction number, and vice versa. A decrease in the reaction numbers implies an increase in the inhibition efficiency of the inhibitor. Both methods reveal that the inhibition efficiency increased with increase in concentration of *Maesobatrya barteri* root extract. An increase in inhibition efficiency with increase in inhibitor concentration indicates a strong interaction between the metal surface and the inhibitor [15].

3.2 Effect of Temperature on Inhibition Efficiency

An increase in temperature led to a decrease in the inhibition efficiency of *Maesobatrya barteri* root extract (Table 2). This indicates a weakening of adsorption bonds between metal surface and inhibitor as well as a physical adsorption mechanism [15].

The activation energy (E_a) of the corrosion process in the absence and presence of the root extract was evaluated using the Arrhenius equation:

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

where CR is the corrosion rate, E_a is the activation energy, R is the universal gas constant, T is the absolute temperature and A is the pre – exponential factor. Hence, a plot of ln CR vs. 1/T should be linear, with a gradient of $-E_a/R$ and an intercept of ln A, if the Arrhenius equation is obeyed. Fig. 3 depicts an Arrhenius plot of ln CR vs. 1/T for aluminium corrosion in 0.5 M HCl solution in the absence and presence of various concentrations of the root extract.

Satisfactory straight lines of high correlation coefficients were obtained. The E_a values were calculated from the slopes of the linear plots and are presented in Table 3. It is clear that E_a values in the presence of various concentrations of root extract are higher than in their absence. An increase in the E_a values in the presence of the extract compared to the blank denotes physical adsorption while the reverse is usually attributed to chemical adsorption [15]. The decrease in the E_a values in the inhibited solution compared to the blank coupled with a decrease in inhibition efficiency as temperature increases supports physical adsorption.

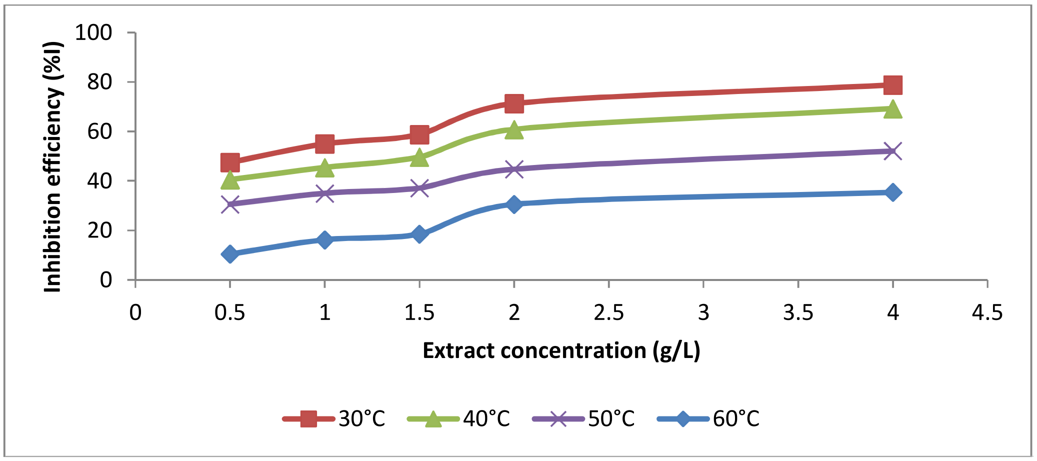

Fig. 1. A plot of inhibition efficiency against various concentrations of *Maesobatrya barteri* root extract at 30℃ - 60℃

C	30℃	40℃	50℃	60℃
0.5	47.5	40.56	30.53	10.48
1	55	45.45	35	16.24
1.5	58.75	49.65	37.11	18.56
2	71.25	60.84	44.74	30.56
4	78.75	69.23	52.11	35.44

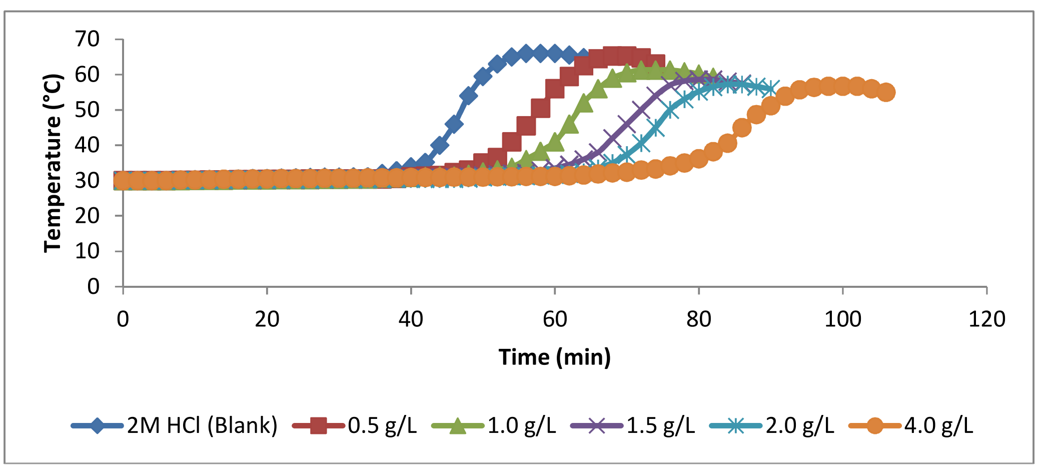

Fig. 2. Temperature – time curves for aluminium corrosion in 2M HCl obtained in absence and presence of *Maesobatrya barteri* root extract

Time (min)	2M HCl (blank)	0.5 g/L	1.0 g/L	1.5 g/L	2.0 g/L	4.0 g/L
0	30	30	30	30	30	30
2	30	30	30	30	30	30
4	30	30	30	30	30	30
6	30	30	30	30	30	30
8	30.2	30.1	30	30.1	30.1	30.2
10	30.3	30.1	30.1	30.1	30.1	30.2
12	30.4	30.2	30.1	30.1	30.1	30.3
14	30.4	30.2	30.1	30.1	30.2	30.4
16	30.5	30.3	30.2	30.1	30.3	30.4
18	30.6	30.3	30.2	30.1	30.3	30.5
20	30.6	30.4	30.2	30.2	30.3	30.5
22	30.7	30.4	30.3	30.2	30.3	30.6
24	30.8	30.4	30.3	30.2	30.3	30.6
26	30.9	30.5	30.3	30.2	30.3	30.6
28	31	30.5	30.4	30.3	30.4	30.7
30	31	30.5	30.4	30.4	30.4	30.7
32	31.1	30.5	30.5	30.4	30.4	30.7
34	31.1	30.5	30.5	30.5	30.4	30.8
36	32	30.5	30.7	30.5	30.4	30.8
38	32.8	30.6	30.8	30.6	30.5	30.8
40	33.9	31.1	30.9	30.7	30.5	30.9
42	35.2	31.3	31.2	30.8	30.5	30.9
44	40	31.5	31.3	30.9	30.5	30.9
46	46	32.3	31.3	31.1	30.5	31
48	54	33	31.8	31.2	30.6	31
50	59.5	35	32.5	31.5	30.9	31
52	63	36.5	33.1	31.8	31.1	31.1
54	65	41	33.8	32	31.2	31.1
56	66	45.5	36	32.2	31.3	31.2
58	66	50.5	38.3	32.8	31.5	31.2
60	66	56	41	33.5	31.6	31.2
62	65.5	59.5	46	34.5	31.9	31.4
64	64.8	62.5	52	36	32.4	31.6
66		64.5	56	38	33.5	31.9
68		65.3	59	42	35	32.2
70		65.3	60.5	46	37.3	32.4
72		64.7	61.3	50	40.6	33
74		63	61.3	54	45	33.3
76			61.3	57	50	34.2
78			60.8	58.2	53	35
80			60.2	58.6	55.2	36.2
82			59.3	58.6	56.5	38.2
84				58	57.2	40.6
86				57.5	57.2	45
88					56.6	48.7
90					55.9	51.2
92						53.9
94						55.7
96						56.4
98						56.7
100						56.7
102						56.7
104						56
106						55

The values of enthalpy of activation (ΔH^0_{ads}) and entropy of activation (ΔS^0_{ads}) were obtained from the transition state equation [18]:

$$CR = \frac{RT}{Nh}\exp\left(\frac{\Delta S^0_{ads}}{R}\right)\exp\left(\frac{-\Delta H^0_{ads}}{RT}\right) \qquad (6)$$

where CR is the corrosion rate, h is the Planck's constant, N is the Avogadro's number, T is the absolute temperature and R is the universal gas constant. A plot of ln (CR/T) vs. 1/T (Fig. 4) was made and straight lines were obtained with slope of ($-\Delta H^0_{ads}/R$) and intercept of [ln (R/Nh) +

$\Delta S^0_{ads}/R$] from which the values of ΔH^0_{ads} and ΔS^0_{ads} were calculated and listed in Table 3. The positive values of ΔH^0_{ads} both in the absence and presence of the root extract reflect the endothermic nature of the aluminium corrosion process while the positive ΔS^0_{ads} values for root extract indicate a system disorder.

3.3 Adsorption Isotherm

The best fit for the adsorption of *Maesobatrya barteri* root extract on aluminium surface was obtained with modified Langmuir adsorption isotherm defined as [19]:

$$\frac{C}{\theta} = \frac{n}{K_{ads}} + nC \quad (7)$$

where C is the inhibitor concentration, θ is the degree of surface while K_{ads} is the equilibrium constant of the adsorption process. Plot of C/θ vs. C gives straight lines (Fig. 5). The values of K_{ads} were evaluated from the intercept of the graph and presented in Table 4. K_{ads} is related to the standard free energy of adsorption (ΔG^0_{ads}) by the formula [20]:

$$K_{ads} = \frac{1}{55.5} exp\left(\frac{-\Delta G^0_{ads}}{RT}\right) \quad (8)$$

where 55.5 is the molar concentration of water in the solution, R is the universal gas constant while T is the absolute temperature.

Table 1. Effect of *Maesobatrya barteri* root extract on inhibition efficiency of aluminium corrosion in 2M HCl solution (thermometric measurements)

Extract Concentration	Minimum Temperature T_i (℃)	Maximum Temperature T_m (℃)	Time taken to reach Maximum temp. t (min)	Reaction Number RN(℃/min)	Inhibition Efficiency (%I)
2M HCl	30.0	66.0	56	0.6429	-
0.5 g/L	30.0	65.3	70	0.5043	21.56
1.0 g/L	30.0	61.3	72	0.4347	32.38
1.5 g/L	30.0	58.6	80	0.3575	44.39
2.0 g/L	30.0	57.2	84	0.3238	49.63
4.0 g/L	30.0	56.7	98	0.2724	57.63

Table 2. Calculated values of weight loss, corrosion rate and inhibition efficiency for aluminium corrosion in 0.5M HCl solution containing *Maesobatrya barteri* root extract at 30℃ – 60℃

Extract Concentra-tion	Weight loss (g)				Corrosion rate (mg cm^{-2} hr^{-1})				Inhibition efficiency (%I)			
	30℃	40℃	50℃	60℃	30℃	40℃	50℃	60℃	30℃	40℃	50℃	60℃
0.5M HCl	0.0080	0.0286	0.0380	0.1250	0.0500	0.1788	0.2375	0.7813	-	-	-	-
0.5 g/L	0.0042	0.0170	0.0264	0.1119	0.0263	0.1063	0.1650	0.6994	47.50	40.56	30.53	10.48
1.0 g/L	0.0036	0.0156	0.0247	0.1047	0.0225	0.0975	0.1544	0.6544	55.00	45.45	35.00	16.24
1.5 g/L	0.0033	0.0144	0.0239	0.1018	0.0206	0.0900	0.1494	0.6363	58.75	49.65	37.11	18.56
2.0 g/L	0.0023	0.0112	0.0210	0.0868	0.0144	0.0700	0.1313	0.5425	71.25	60.84	44.74	30.56
4.0 g/L	0.0017	0.0088	0.0182	0.0807	0.0106	0.0538	0.1138	0.5044	78.75	69.23	52.11	35.44

Table 3. Calculated values of thermodynamic parameters for aluminium corrosion in 0.5 M HCl solution containing *Maesobatrya barteri* root extract

Extract concentration	E_a (kJ mol^{-1})	ΔH^0_{ads}(kJ mol^{-1})	ΔS^0_{ads}(J K^{-1} mol^{-1})
0.5M HCl (Blank)	72.0549	69.4186	-40.2472
0.5 g/L	86.6984	84.0629	2.4285
1.0 g/L	89.1593	86.5165	9.3875
1.5 g/L	91.0466	88.4111	14.8579
2.0 g/L	97.0826	94.4470	31.9931
4.0 g/L	103.9250	101.2811	51.8885

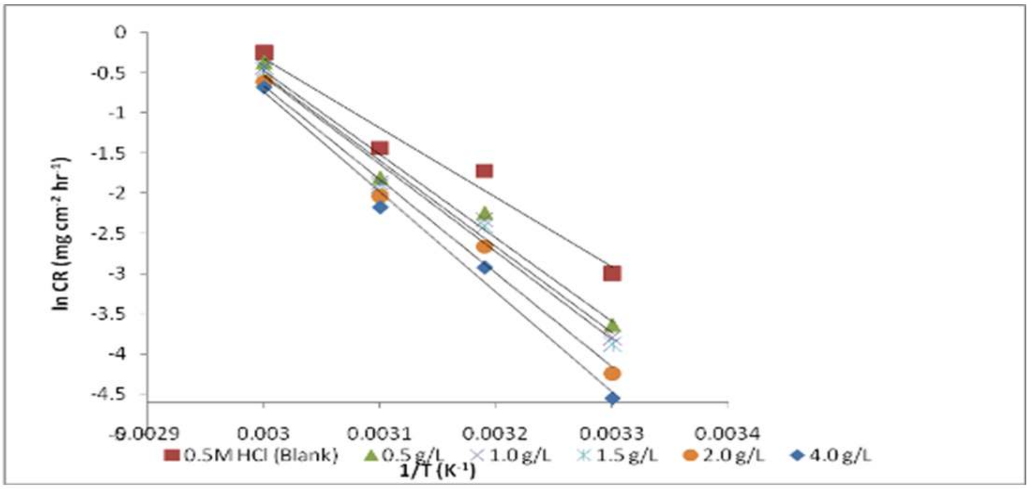

Fig. 3. Plot of ln CR vs. 1/T (Arrhenius plot) for aluminium corrosion in 0.5M HCl in the absence and presence of *Maesobatrya barteri* root extract

1/T	ln CR 0.5M (Blank)	0.5 g/L	1.0 g/L	1.5 g/L	2.0 g/L	4.0 g/L
0.003	-0.2468	-0.3575	-0.424	-0.4521	-0.6116	-0.6844
0.0031	-1.4376	-1.8018	-1.8682	-1.9011	-2.0303	-2.1733
0.00319	-1.7215	-2.2415	-2.3279	-2.4079	-2.6593	-2.9225
0.0033	-2.9957	-3.6382	-3.7942	-3.8825	-4.2405	-4.5469

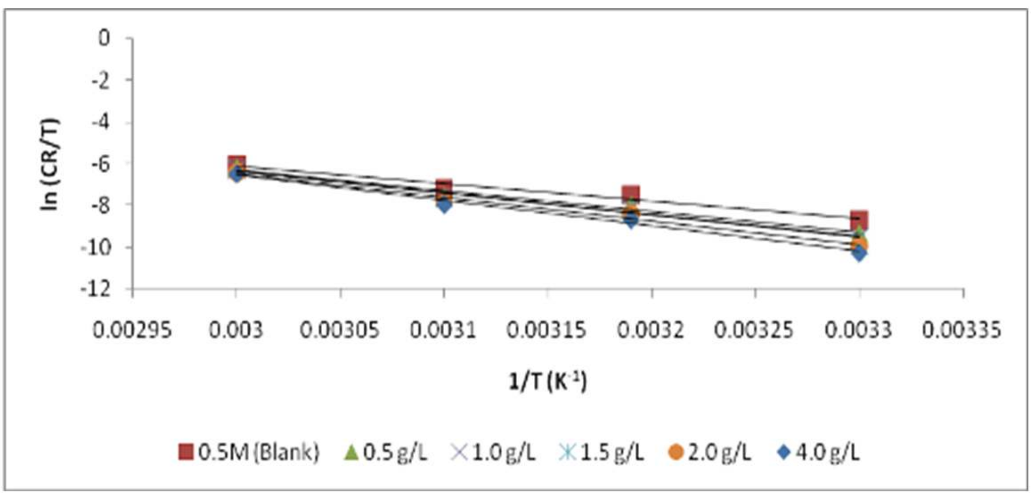

Fig. 4. Plot of ln (CR/T) vs 1/T (Transition state plot) for aluminium corrosion in 0.5M HCl solution in the absence and presence of *Maesobatrya barteri* root extract

1/T	ln (CR/T) 0.5M HCl (Blank)	0.5 g/L	1.0 g/L	1.5 g/L	2.0 g/L	4.0 g/L
0.003	-6.0549	-6.1657	-6.2322	-6.2602	-6.4197	-6.4925
0.0031	-7.2152	-7.5795	-7.6459	-7.6788	-7.8079	-7.951
0.00319	-7.4677	-7.9877	-8.0741	-8.1541	-8.4055	-8.6687
0.0033	-8.7095	-9.3519	-9.508	-9.5962	-9.9543	-10.2606

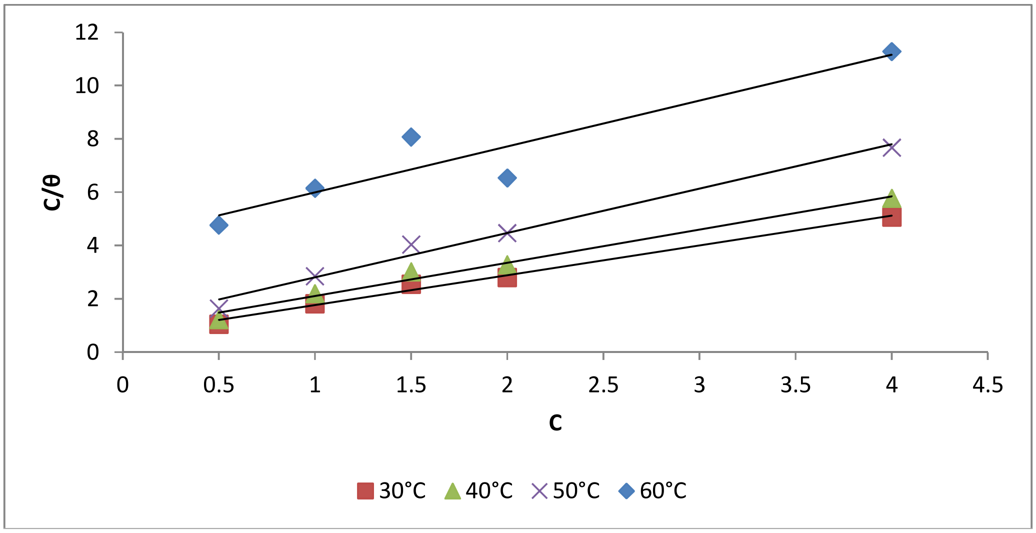

Fig. 5. Plot of C/θ vs. C (Langmuir isotherm) for aluminium corrosion in 0.5M HCl solution containing *Maesobatrya barteri* root extract

C	C/θ 30℃	40℃	50℃	60℃
0.5	1.0526	1.2327	1.6377	4.771
1	1.8182	2.2002	2.8571	6.1576
1.5	2.5532	3.0211	4.042	8.0819
2	2.807	3.2873	4.4703	6.5445
4	5.0794	5.7778	7.6761	11.2867

Table 4. Some parameters of the linear regression of langmuir adsorption isotherm for aluminium corrosion in 0.5M HCl solution containing *Maesobatrya barteri* root extract

Temperature	R^2	n	K_{ads} (g^{-1} L)	ΔG^0_{ads} (kJ mol^{-1})
30℃	0.9906	1.12	1.7088	-11.4676
40℃	0.9856	1.25	1.4493	-11.4174
50℃	0.9857	1.67	1.4609	-11.8036
60℃	0.8767	1.72	0.4043	-8.6124

The thermodynamic parameters for the adsorption of *Maesobatrya barteri* root extract on aluminium surface are shown in Table 4. The linear plots have 'n' values (gradients) greater than 1, indicating that the extract occupied more than one adsorption site on the metal surface. The negative values of ΔG^0_{ads} indicate the spontaneity of the adsorption process. Additionally, the values of ΔG^0_{ads} in this work being less than -40kJ mol^{-1}, the threshold value for chemisorption, further support the claim that the adsorption mode by the root extract was by physisorption.

4. CONCLUSION

The following conclusions can be drawn from this study: *Maesobatrya barteri* root extract significantly inhibited the corrosion of aluminium in HCl solutions at 30℃ – 60℃, showing that the extract is a good inhibitor of aluminium corrosion in HCl solution. The inhibition efficiency increased with increase in extract concentration but decreased with increase in temperature. An increase in inhibition efficiency with increase in the extract concentration reveals a strong adsorption of the inhibitor on aluminium surface thereby reducing its rate of corrosion while a decrease in the inhibition efficiency with increase in temperature indicates a weakening of adsorption bonds between the metal surface and the inhibitor, as temperature increases. The calculated values of ΔH^0_{ads} for aluminium corrosion in 0.5 M HCl being positive indicate the endothermic nature of the adsorption process while the negative values of ΔG^0_{ads} obtained reveal the spontaneity of the inhibition process. The increase in the E_a values in the presence of the extract compared to the blank coupled with a decrease in the inhibition efficiency with increase in temperature indicates physical adsorption mechanism. Adsorption of *Maesobatrya barteri* root extract on aluminium surface obeyed the modified Langmuir adsorption isotherm.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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