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Solenostemon monostachyus Leaf Extract as Eco-friendly Inhibitor of Aluminium Corrosion in Acidic Medium

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Author's contribution

The sole author designed, analyzed and interpreted and prepared the manuscript.

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ABSTRACT

The inhibitory effect of *Solenostemon monostachyus* leaf extract on aluminium corrosion in hydrochloric acid solution was studied using weight loss, thermometric and hydrogen evolution methods. The results obtained reveal that *Solenostemon monostachyus* leaf extract functioned as a good inhibitor of aluminium corrosion in HCl solution. Inhibition efficiency was found to increase with increase in extract concentration and temperature. Chemical adsorption mechanism has been proposed for the adsorption of the leaf extract onto aluminium surface. Thermodynamic parameters revealed that the adsorption of the extract onto the metal surface was endothermic and spontaneous. The adsorption of *Solenostemon monostachyus* leaf extract on aluminium surface obeyed the Freundlich adsorption isotherm.

Keywords: Corrosion inhibition; Solenostemon monostachyus; aluminium; Freundlich isotherm; chemisorptions; weight loss; thermometric; hydrogen evolution.

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1. INTRODUCTION

Aluminium has a wide range of domestic and industrial applications. The corrosion resistance of aluminium in aggressive media is very good and is attributed to a thin, oxide film on its surface. This oxide film is amphoteric and dissolves in high concentrations of acids and bases [1]. Consequently, corrosion inhibitors are required for the protection of aluminium in such environments.

The desire for eco-friendly inhibitors of aluminium corrosion has yielded positive results, with the extraction of efficient inhibitors from natural products, especially plants. Plant extracts are renewable, cheap, non-toxic, and environmentally friendly. Several leaves extracts have been reported as good inhibitors of aluminium corrosion in acidic media [2-6]. The search for efficient eco-friendly inhibitors of aluminium corrosion is ongoing because none of the reported inhibitors offers a 100% protection to aluminium in acidic medium.

Solenostemon monostachyus (English name: Monkey potato; Ibibio name: Ntorikwot; Yoruba name: Olojogbodu) is an edible medicinal plant belonging to the family Lamiaceae. It is a slightly succulent, aromatic plant that grows up to 100 cm tall [7]. The traditional medicinal uses of *Solenostemon monostachyus* plant by the people of Nigeria have been reported [8-10]. The phytochemical analysis of *Solenostemon monostachyus* leaf extract revealed the presence of tannins, saponins, alkaloids, flavonoids and cyanogenic glycosides [11].

In furtherance of efforts geared at broadening the database of efficient eco-friendly corrosion inhibitors, the aim of this work was to assess the inhibitory effect of *Solenostemon monostachyus* leaf extract on aluminium corrosion in hydrochloric acid solution. There is no reported work on the inhibition of aluminium corrosion by *Solenostemon monostachyus* leaf extract in acidic medium.

2. MATERIALS AND METHODS

2.1 Test Materials

Aluminium sheet used for this work was obtained from System Metal Industries Limited, Calabar, Nigeria. It had the following chemical composition (weight%): Al (99.16), Fe (0.35), Si (0.25), Cu (0.05), Zn (0.05), V (0.05), Ti (0.03), Mn (0.03) and Mg (0.03). Each sheet was 0.04 cm in thickness and was mechanically press cut into 5 cm x 4 cm coupons. These coupons were polished to mirror finish using different grades of silicon carbide papers. The coupons were degreased in absolute ethanol, dried in acetone and stored in a moisture – free desiccator before use in corrosion studies.

2.2 Preparation of *Solenostemon monostachyus* Leaf Extract

Fresh leaves of *Solenostemon monostachyus* were collected from a farm in Etinan, Akwa Ibom State, Nigeria and authenticated by a plant taxonomist in the Department of Botany and Ecological Studies, University of Uyo, Nigeria. They were plucked, washed and oven-dried at 50°C to a constant weight. They were then ground to powder. Stock solution of *Solenostemon monostachyus* leaf extract was prepared by soaking 5.0 g of the dried and ground leaves in 1.0 L of 0.5 M HCl solution. The resultant solution was kept for 24 hours, filtered and stored. From the stock solution (5.0 g/L), extract concentrations of 1.0 g/L, 2.0 g/L, 3.0 g/L, and 4.0 g/L, respectively, in 0.5 M HCl solution were prepared for the weight loss studies at 30°C, 40°C, 50°C, and 60°C. The same extract concentrations were prepared in 2 M HCl solution for the hydrogen evolution and thermometric tests.

2.3 Weight Loss Method

Previously weighed aluminium coupons were suspended with the aid of glass hooks and rods and immersed in 100 cm^3 of 0.5 M HCl solution (blank) and in 0.5 M HCl solution containing 1.0 g/L – 4.0 g/L *Solenostemon monostachyus* leaf extract (inhibitor) in open beakers. In each experiment, one aluminium coupon per beaker was used. The beakers were then placed in a thermostatic water bath maintained at 30°C, 40°C, 50°C, and 60°C, respectively. The aluminium coupons were retrieved from the test solutions after four (4) hours and scrubbed with bristle brush under running water. They were dipped in acetone and air - dried before reweighing. The weight loss was recorded.

The inhibition efficiency I(%) was calculated using the formula [12]:

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

where W_0 and W_1 are the weight losses of the aluminium coupons in the absence and presence of extract, respectively, in 0.5 M HCl at the same temperature.

The degree of surface coverage (θ) was obtained by the equation:

$$\theta = \left(1 - \frac{W_1}{W_0}\right) \quad (2)$$

where W_0 and W_1 are the weight losses of the aluminium coupons in the absence and presence of extract, respectively, in 0.5 M HCl at the same temperature.

The corrosion rate (CR) was calculated using the formula [2]:

$$\mathrm{CR}\ (\mathrm{mg\ cm^{-2} hr^{-1}}) = \left(\frac{W}{At}\right) \quad (3)$$

where W is the weight loss (mg), A is the total surface area (cm^2) while t is the exposure time (hr).

2.4 Hydrogen Evolution Method

The reaction vessel and procedure for measuring the corrosion process by this method are as described in literature [13]. A 100 cm^3 of 2 M HCl solution was introduced into the reaction vessel connected to a burette through a delivery tube. The initial volume of air in the burette was recorded. One 5 cm x 4 cm aluminium coupon was dropped into the 2 M HCl solution and the reaction vessel quickly closed to prevent any escape of hydrogen gas. The volume of H_2 gas evolved from the corrosion reaction was monitored by the depression (in cm^3) in the paraffin oil level. The depression in paraffin oil level was monitored every 60 seconds for 20 minutes. The same experiment was repeated in the presence of 1.0 g/L – 4.0 g/L Solenostemon monostachyus leaf extract in 2 M HCl solution.

The inhibition efficiency I(%) was calculated using the equation:

$$\mathrm{I}(\%) = \left(1 - \frac{R_{Hi}}{R_{H0}}\right) \times 100 \quad (4)$$

where RH_o and RH_i are the rates of evolution of H_2 gas in the absence and presence of inhibitor, respectively, at a specified time.

2.5 Thermometric Method

The reaction vessel and procedure for determining the corrosion behaviour by this method has been described by other workers [14–15]. The reaction vessel was insulated with cotton wool to prevent heat loss. In the thermometric technique, the corrodent concentration was kept at 2 M HCl. The volume of test solution used was 50 cm^3. The initial temperature in all experiments was kept at 30.0°C. The progress of corrosion reaction was monitored by determining the changes in temperature with time using a calibrated thermometer (0-100°C) to the nearest ± 0.1°C. This method enabled the computation of the reaction number (RN) defined as [15]:

$$\mathrm{RN}\ (^\circ\mathrm{C\ min^{-1}}) = \frac{T \quad - T_i}{t} \quad (5)$$

where T_m and T_i are the maximum and initial temperatures, respectively, while 't' is the time (min) taken to reach the maximum temperature. The inhibition efficiency, I(%) was evaluated from the percentage reduction in the reaction number via the equation:

$$\mathrm{I}(\%) = \left(1 - \frac{RN_1}{RN_0}\right) \times 100 \quad (6)$$

where RN_0 is the reaction number in the absence of inhibitor (blank) and RN_1 is the reaction number in the presence of extract (inhibitor).

3. RESULTS AND DISCUSSION

3.1 Effect of *Solenostemon monostachyus* Leaf Extract Concentration on Inhibition Efficiency

Fig. 1 shows the effect of *Solenostemon monostachyus* leaf extract concentration on the inhibition efficiency of aluminium corrosion in 0.5 M HCl solution by weight loss measurements. It is observed that the inhibition efficiency increased with increase in the leaf extract concentration, at a particular temperature. The effect of *Solenostemon monostachyus* leaf extract on the volume of H_2 gas evolved in the corrosion of aluminium in 2 M HCl is depicted in Fig. 2. Fig. 2 reveals that as the leaf extract concentration increases, the volume of H_2 gas evolved decreases. The calculated values of inhibition efficiency for the inhibition process containing *Solenostemon monostachyus* leaf extract are presented in Table 1. Table 1 reveals that the inhibition efficiency increased with increase in the concentration of

Solenostemon monostachyus leaf extract. Fig. 3 depicts the thermometric results for aluminium corrosion in 2 M HCl solution containing *Solenostemon monostachyus* leaf extract. It is observed that as the concentration of the leaf extract increases, the time required to reach the maximum temperature (t) increases while the maximum temperature (T_m) decreases. Table 2 contains the calculated values of reaction number (RN) and inhibition efficiency I(%) for aluminium corrosion in 2 M HCl containing *Solenostemon monostachyus* leaf extract. Table 2 shows an increase in the inhibition efficiency with increase in the extract concentration. It is instructive to note that the inhibition efficiencies obtained by the weight loss, hydrogen evolution and thermometric methods were high and followed a similar trend. This confirms that *Solenostemon monostachyus* leaf extract is a good inhibitor of aluminium corrosion in HCl solution. Furthermore, an increase in the inhibition efficiency with increase in extract concentration indicates that the extract inhibits the corrosion process by adsorbing on the metal surface, forming protective thin films which reduce/stop the electron transfer process on the metal surface [16].

3.2 Effect of Temperature on inhibition Efficiency

Table 3 shows that the inhibition efficiency increased with increase in temperature at 1.0 g/L –4.0 g/L *Solenostemon monostachyus* leaf extract concentrations studied. An increase in inhibition efficiency with increase in temperature indicates a strong interaction between the metal surface and inhibitor as well as a chemical adsorption mechanism. Consequently, *Solenostemon monostachyus* leaf extract chemically adsorbed on the aluminium surface.

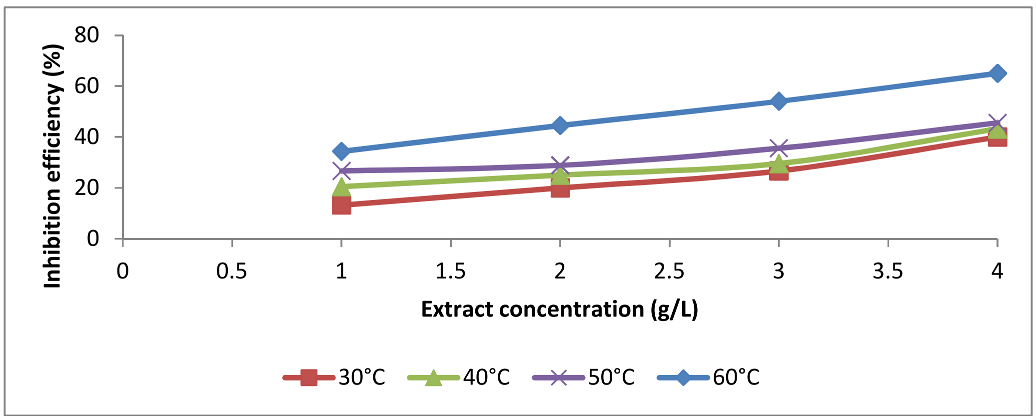

Fig. 1. Effect of *Solenostemon monostachyus* leaf extract concentration on the inhibition efficiency of aluminium corrosion in 0.5 M HCl solution at different temperatures

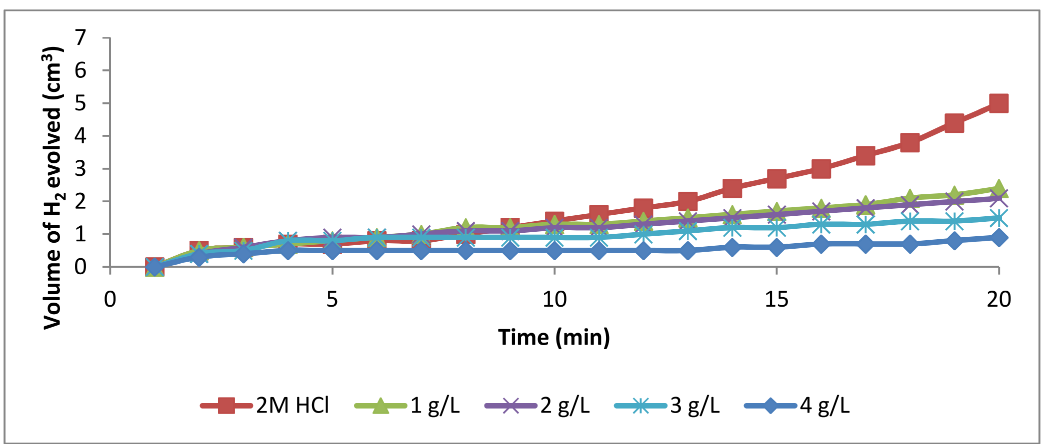

Fig. 2. Variation of volume of H_2 gas evolved (cm^3) with time (min) for aluminium corrosion in 2 M HCl in the absence and presence of *Solenostemon monostachyus* leaf extract at 30°C

Table 1. Effect of *Solenostemon monostachyus* leaf extract concentration on inhibition efficiency of aluminium in 2 M HCl solution at 30°C (Hydrogen evolution measurements)

Extract concentration (g/L)	H_2 evolution rate (cm^3 min^{-1})	Inhibition efficiency (%)
Blank	0.29	-
1.0	0.14	51.72
2.0	0.12	58.62
3.0	0.08	72.41
4.0	0.05	82.76

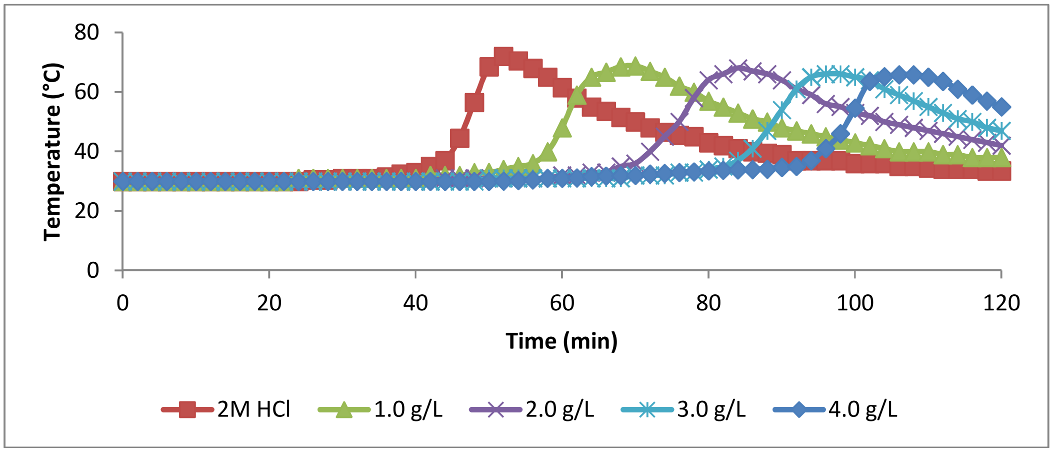

Fig. 3. Temperature – time curves for aluminium corrosion in 2 M HCl obtained in absence and presence of *Solenostemon monostachyus* leaf extract

Table 2. Effect of *Solenostemon monostachyus* leaf extract on inhibition efficiency of aluminium corrosion in 2 M HCl solution (Thermometric measurements)

Extract concentration (g L^{-1})	Initial temperature T_i (°C)	Maximum temperature T_m (°C)	Time taken to reach max. temp. t (min)	Reaction number RN (°C min^{-1})	Inhibition efficiency (%)
Blank	30.0	72.0	52	0.8077	-
1.0	30.0	68.8	70	0.5543	31.37
2.0	30.0	68.0	85	0.4471	44.65
3.0	30.0	66.0	95	0.3789	53.09
4.0	30.0	65.8	106	0.3377	58.19

Table 3. Calculated values of weight loss, corrosion rate and inhibition efficiency for aluminium corrosion in 0.5 M HCl solution containing *Solenostemon monostachyus* leaf extract at 30°C – 60°C

Extract conc.	Weight loss (g)				Corrosion rate (mg cm^{-2} hr^{-1})				Inhibition efficiency(%)			
	30°C	40°C	50°C	60°C	30°C	40°C	50°C	60°C	30°C	40°C	50°C	60°C
Blank	0.015	0.044	0.090	0.346	0.094	0.275	0.563	2.163	-	-	-	-
1.0 g/L	0.013	0.035	0.066	0.227	0.081	0.219	0.413	1.419	13.33	20.45	26.67	34.39
2.0 g/L	0.012	0.033	0.064	0.192	0.075	0.206	0.400	1.200	20.00	25.00	28.89	44.51
3.0 g/L	0.011	0.031	0.058	0.159	0.069	0.194	0.363	0.994	26.67	29.55	35.56	54.05
4.0 g/L	0.009	0.025	0.049	0.151	0.056	0.156	0.306	0.756	40.00	43.18	45.56	65.03

The values of the activation energy (E_a) for aluminium corrosion in 0.5 M HCl solution in the absence and presence of *Solenostemon monostachyus* leaf extract, respectively, were obtained using the alternative formulation of Arrhenius equation [16]:

$$\ln CR = \frac{-E_a}{RT} + \ln A \qquad (7)$$

Where CR is the corrosion rate, R is the universal gas constant, T is the absolute temperature while A is the pre-exponential factor.

The activation energies (E_a) of aluminium corrosion in 0.5 M HCl solution, with and without inhibitors, were obtained from the gradients of ln CR vs. 1/T plots (Fig. 4) and the results presented in Table 4. Table 4 shows that the E_a values in the presence of the leaf extract were lower than the E_a value of the blank (84.23 kJ mol^{-1}). The decrease in the E_a values in the presence of the extract indicates chemical adsorption while the reverse signifies physical adsorption [17]. Consequently, *Solenostemon monostachyus* leaf extract adsorbed chemically onto aluminium surface.

The values of enthalpy of activation (ΔH°_{ads}) and entropy of activation (ΔS°_{ads}) were obtained from an alternative formulation of the transition state equation [12]:

$$\ln\left(\frac{CR}{T}\right) = \left[\ln\left(\frac{R}{Nh}\right) + \frac{\Delta S^\circ_{ads}}{R}\right] - \frac{\Delta H^\circ_{ads}}{RT} \qquad (8)$$

where CR is the corrosion rate, T is the absolute temperature, A is the Arrhenius pre-exponential factor, R is the universal gas constant, h is the Planck's constant, and N is the Avogadro's number. Fig. 5 shows linear plots of ln (CR/T) vs. 1/T with gradients of ($-\Delta H^\circ_{ads}/R$) and intercepts of [ln (R/Nh) + $\Delta S^\circ_{ads}/R$] from which the values of ΔH°_{ads} and ΔS°_{ads} were calculated and listed in Table 4. The positive values of ΔH°_{ads} both in the blank and in the presence of extracts indicate the endothermic nature of the aluminium corrosion process.

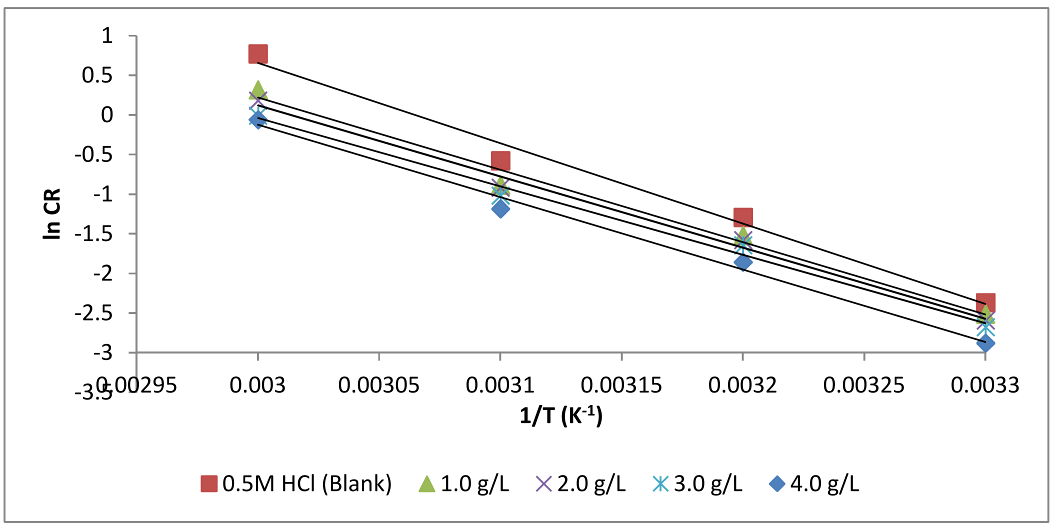

Fig. 4. Plot of ln CR vs. 1/T (Arrhenius plot) for aluminium corrosion in 0.5 M HCl in the absence and presence of *Solenostemon monostachyus* leaf extract

Table 4. Calculated values of thermodynamic parameters for aluminium corrosion in 0.5 M HCl solution in the absence and presence of *Solenostemon monostachyus* leaf extract

Extract concentration	E_a (kJ mol^{-1})	ΔH°_{ads} (kJ mol^{-1})	ΔS°_{ads} (J K^{-1} mol^{-1})
0.5 M HCl (Blank)	84.23	81.57	4.33
1.0 g/L	76.61	73.99	- 21.86
2.0 g/L	74.66	72.05	- 28.69
3.0 g/L	71.83	69.21	- 38.52
4.0 g/L	70.41	67.79	- 44.77

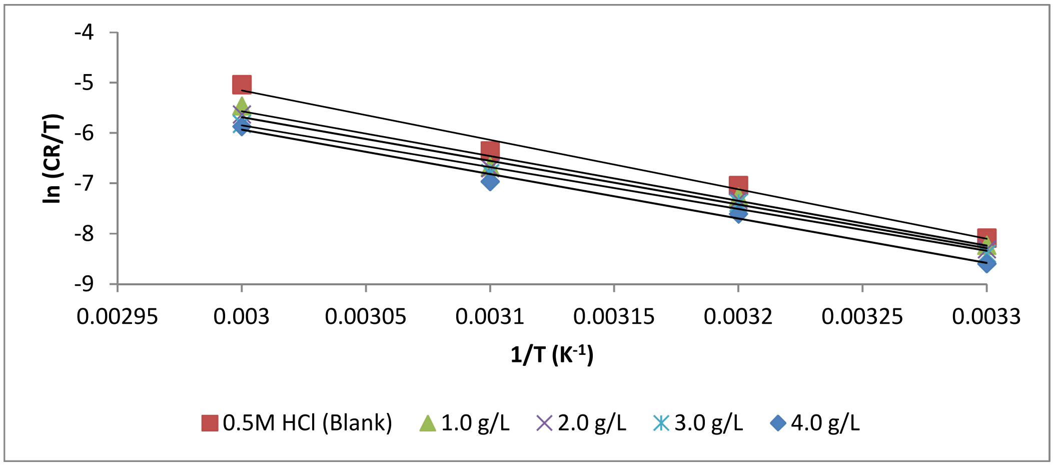

Fig. 5. Plot of ln (CR/T) vs. 1/T (Transition state plot) for aluminium corrosion in 0.5 M HCl solution in the absence and presence of *Solenostemon monostachyus* leaf extract

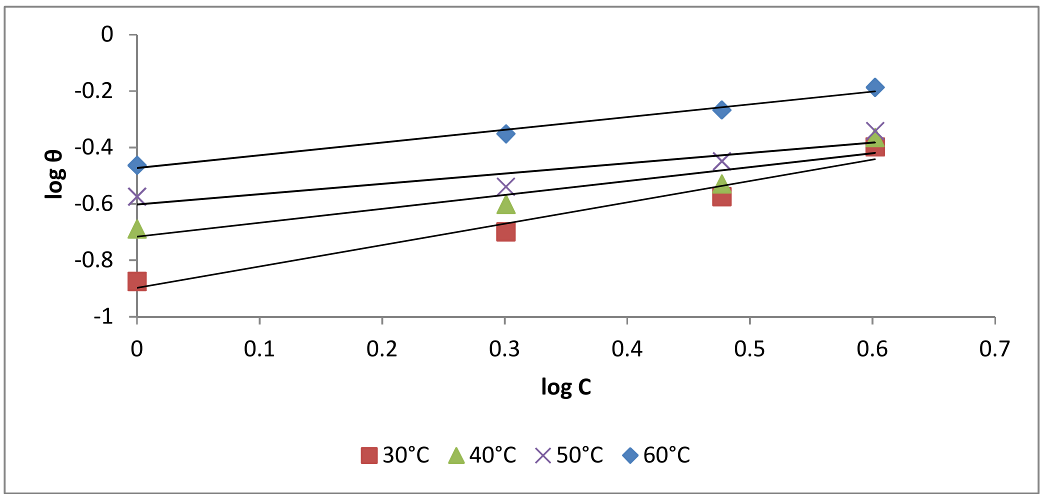

Fig. 6. Variation of log θ vs. log C (Freundlich isotherm) for aluminium corrosion in 0.5 M HCl solution containing *Solenostemon monostachyus* leaf extract at different temperatures

Table 5. Some parameters of the linear regression of Freundlich adsorption isotherm for aluminium corrosion in 0.5 M HCl solution containing *Solenostemon monostachyus* leaf extract

Temperature	R^2	n	log K_{ads}	K_{ads}	$\Delta G°_{ads}$ (kJ mol^{-1})
303K	0.9614	0.76	-8.98×10^{-1}	1.27×10^{-1}	-4.92
313K	0.8728	0.49	-7.16×10^{-1}	1.92×10^{-1}	-6.16
323K	0.8422	0.37	-6.02×10^{-1}	2.50×10^{-1}	-7.06
333K	0.9860	0.45	-4.72×10^{-1}	3.37×10^{-1}	-8.13

3.3 Adsorption Isotherm

The adsorption of *Solenostemon monostachyus* leaf extract obeyed the Freundlich adsorption isotherm equation [18]:

$$\log \theta = n \log C + \log K_{ads} \quad (9)$$

where θ is the degree of surface coverage, K_{ads} is the equilibrium adsorption constant, C is the inhibitor concentration and n is the interaction parameter. Fig. 6 (above) reveals linear plots of log θ vs. log C, with gradients of 'n' and intercepts of log K_{ads} showing that the Freundlich adsorption isotherm is obeyed. The equilibrium adsorption constant, K_{ads}, is related to the standard free energy of adsorption ($\Delta G°_{ads}$) by the formula [19,20]:

$$K_{ads} = \frac{1}{55.5}\exp\left(\frac{-\Delta G^{\circ}_{ads}}{RT}\right) \quad (10)$$

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

Some parameters of the linear regression of Freundlich adsorption isotherm are contained in Table 5. The negative values of $\Delta G°_{ads}$ obtained indicate the spontaneity of the adsorption process. Additionally, the values of K_{ads} obtained increased with increase in temperature. An increase in the value of K_{ads} with increase in temperature indicates that the extract adsorbed more strongly onto aluminium surface with increase in temperature [12]. This proposition is supported by an increase in the inhibition efficiency of *Solenostemon monostachyus* leaf extract with increase in temperature.

4. CONCLUSION

On the basis of this study, the following conclusions could be drawn: *Solenostemon monostachyus* leaf extract is a good inhibitor of aluminium corrosion in hydrochloric acid solution. The inhibition efficiency increased with increase in *Solenostemon monostachyus* leaf extract concentration and temperature. The calculated thermodynamic parameters reveal that the adsorption of *Solenostemon monostachyus* leaf extract onto aluminium surface was spontaneous and endothermic. Based on an increase in the inhibition efficiency with increase in temperature coupled with a decrease in the E_a value in the extract compared to the blank, chemical adsorption has been proposed for the adsorption of the leaf extract onto aluminium surface. An increase in the value of equilibrium adsorption constant (K_{ads}) with increase in temperature indicates that the extract adsorbed more strongly onto aluminium surface with increase in temperature. The adsorption of *Solenostemon monostachyus* leaf extract obeyed the Freundlich adsorption isotherm.

COMPETING INTERESTS

Author has declared that no competing interests exist.

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