Corrosion Inhibition and Adsorption Characteristics of *Jatropha curcas* Leaves Extract on Aluminium in 1M HCl

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Abstract   Corrosion inhibition of aluminium (AA 1060) in 1M HCl by *Jatropha curcas* leave extracts was studied using weight loss method at 30°C, 40°C, 50°C and 60°C. The results obtained indicate that the extracts inhibited the corrosion process in the medium by adsorption and that corrosion rate increased with rise in temperature. Corrosion rate was also found to decrease in the presence of the inhibitor as compared to the blank. Inhibition efficiency was found to increase with increased inhibitor concentration but reduces with increase in temperature to a maximum of 76.49% at 30°C and 0.5g/L concentration of the leave extract. The change in Gibbs free energy of adsorption was -16.99kJ/mol at 30°C and decrease with increase in temperature to -13.54kJ/mol at 60°C. The adsorption of the plant extract on aluminium surface was found to obey Temkin, Freundlich and El-Awady adsorption isotherms. Physical adsorption mechanism is proposed for the adsorption of the leaves extract on the aluminium surface.

Keywords   *Jatropha Curcas*, Heat of Adsorption, Corrosion Inhibition, Physical Adsorption

1. Introduction

Aluminium, Al is a metallic material that has properties that allow quite different uses and somewhat opposites. Due to its resistance to corrosion in many media and their mechanical properties, reflected in a low density and good mechanical tensile strength, it is a material with excellent characteristics for use in construction and aerospace industry [1]. One of the most important applications of Al and its alloys is found in aluminium-air technology, which was of particular interest for its application to electric vehicle propulsion as well as its low production cost and the existence of a large base for manufacture and distribution [2].

Acid solutions have significant application in industrial processes which includes acid cleaning, descaling and pickling of metallic structures, oil well acidification, electroplating and electro polishing which are usually accompanied by considerable dissolutions of the metal. A useful method of protecting metals like Al and alloys deployed in service in aggressive environments against corrosion is addition of species to the solution in contact with the surface in order to inhibit the corrosion reaction and hence reduce the corrosion rate [3, 4]. Adsorption depends mainly on the charge and nature of the metal surface, electronic characteristics of the metal surface, adsorption of solvent and other ionic species, temperature of corrosion reaction and on the electrochemical potential at solution interface and the type of electrolyte [5]. Originally, inorganic inhibitors such as zinc chromate, polyphosphate and nitrite were used as inhibitors. However, discharge of such material has become unacceptable due to their toxicity and environmental hazards [6]. Accordingly therefore, greater research efforts have been directed towards formulating environmentally acceptable inhibitors. The efficacy of plant extract to act as a successful corrosion inhibitor is its ability to get adsorbed on the surface of the metal [7]. Therefore, the corrosion rate in the presence of the inhibitor may be taken to represent the number of potentially corroding sites that remain after blockage via the plant extract adsorption. It has been observed [8] that the extent and mode of adsorption of corrosion inhibitors on the metal surface depends mainly on certain physico-chemical properties of the molecule such as functional groups, aromaticity, steric factors, electron density at the donor atoms and π orbital character of the donating electrons, and also on the electronic structure of the molecules.

Recent awareness emphasizes that plant products containing compounds like tannins, alkaloids, saponnins, essential oils, flavonoids, organic and amino acids are known to exhibit corrosion inhibiting action for steel and aluminium in acidic environment [9-14]. Such organic compounds are known to contain oxygen, nitrogen,

phosphorus, and sulphur, in a conjugated system [8] and they inhibit via adsorption of the molecule on the metal surface thereby blocking the active corrosion sites [15]. The extract from plant leaves, roots and seeds have been studied and found to be cheap, environmentally friendly and good corrosion inhibitors. Among the plant leave extract studied include opuntia extract [16], carica papaya and *Azadirachta indica* [17], *Vernonia amygdalina* [18], *Telfaria occidentalis* [19], *Phyllanthus amarus* [20], fenugreek leaves [21], olive leaves [22], Euphorbia hirta and *Dialum guieense* [23]. Among the roots studied are ginseng [24], Beet [25], *Kopsia singapurensis* extract [26] and *Murraya koenighii* [27] bamboo leaf extract [28].

In the light of the trend in the application of environmentally benign materials as corrosion inhibitors, we report in this study the adsorptive behaviour and evaluation of the corrosion inhibition performance of Jatropha curcas leaves extract. Jatropha curcas is a flowering/ornamental plant. It is a hedge plant and live-fence used to demarcate farms especially in Northern Africa and South Eastern Nigeria. The oil and seeds are used as purgative to cure ring worm, craw-craw, skin infections, rheumatic pains, dropsy and paralysis by rubbing on the affected parts [29]. The leaves have also been shown to effectively act as a sensitizer in Gratzel Cells [30].

2. Experimental Procedure

Materials: Aluminium sheets of type AA 1060 with 98% purity was purchased at system metals industries limited Calabar, Nigeria. The sheets were mechanically press-cut into coupons of dimension 5cm x 4cm with the thickness of 0.14cm. They were degreased by washing in absolute ethanol and the surface polished with emery paper. The coupons were then re-washed in absolute ethanol, rinsed in acetone to dry and stored in a moisture-free desiccator prior to use. All reagents used for the study were of BDH analytical grade.

Plant Extract: The leaves of *Jatropha curcas* were obtained from Afaha Offiong in Nsit Ibom Local Government Area of Akwa Ibom State, Nigeria. The leaves were sun-dried and ground to powder and 300g of the powdered leaves weighed and soaked in 96% ethanol in a glass column. After 72 hours the extract was ran out of the column and concentrated to paste using the rotatory evaporator. Various masses of the inhibitor, ranging from 0.1g – 0.5g were weighed and dissolved in 1dm^3 of 1M HCl.

Weight Loss Method: In weight loss method, previously weighed aluminium coupons were immersed in 100ml of 1M HCl solution in an open beaker. In each experiment, the cleaned aluminium coupons was weighed and suspended with the aid of glass rod and hook in a beaker containing 100ml acid solution. The beaker was placed in a water bath maintained at temperatures of 30, 40, 50 and 60 ℃ respectively. The procedures were conducted with and without various concentrations of the inhibitors. After 8h, the aluminium coupons were withdrawn from the test solution and immersed in 70% nitric acid for 3 minutes at room temperature. They were scrubbed with a bristle brush under running water, dried in acetone and re-weighed. The weight loss was taken as the difference in weight of the coupon before and after immersion determined by a digital weighing balance. The test was conducted in triplicate to guarantee reliability of the result and the mean value of the weight loss was reported.

3. Result and Discussion

Corrosion Rate and Inhibition Efficiency: Assessment of corrosion rate for aluminium in the different inhibitor/corroding solution was carried out using Eqn. 1 [28].

$$\nu = W_1 - W_2 \, / \, At \qquad (1)$$

where ν is corrosion rate, W_1 and W_2 are the weight (g) before and after immersion in the test solutions, A is the area of the specimen (cm^2) and t is the exposure time (h).
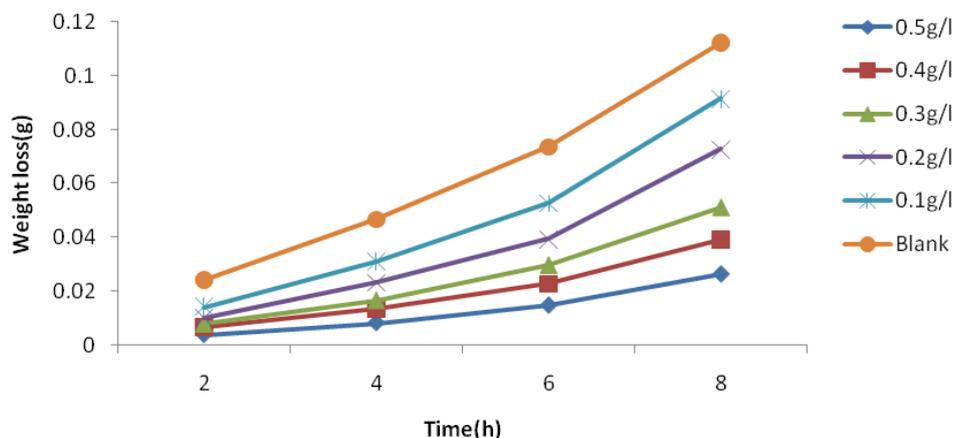

Figure 1. Plot of weight loss against time for Al corrosion in 1M HCl with JCLE at 30°C
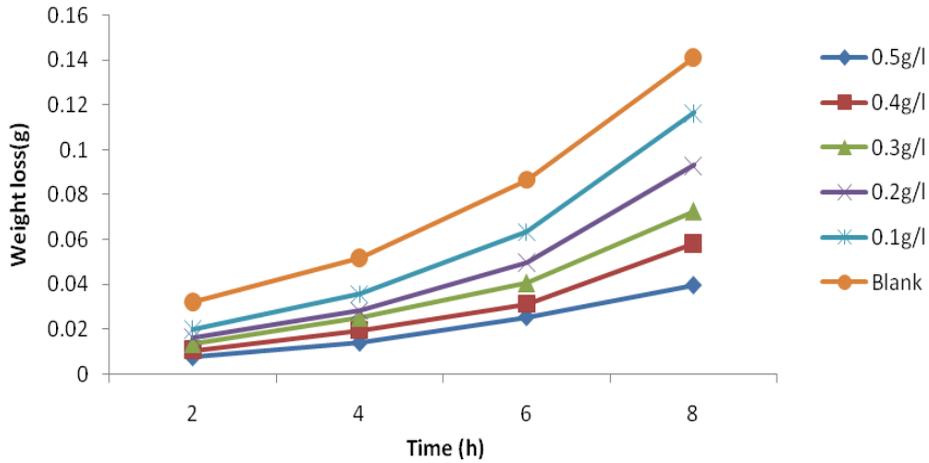
Figure 2. Plot of weight loss against time for Al corrosion in 1M HCl with JCLE at 40℃
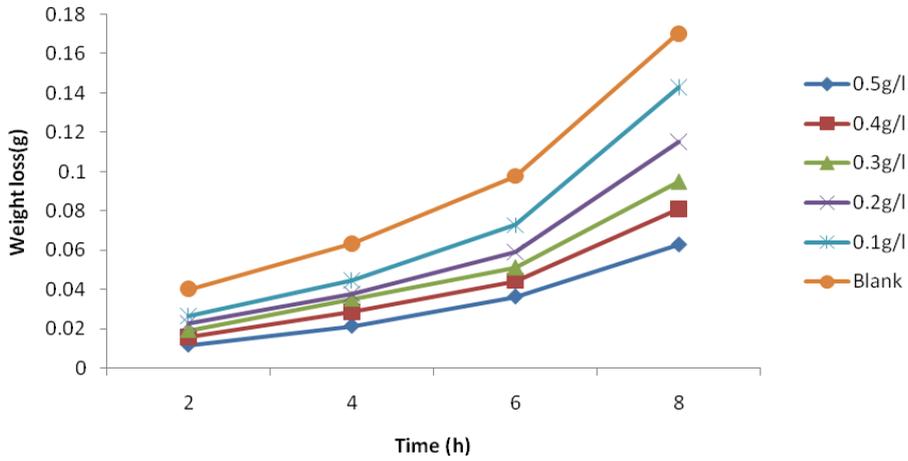

Figure 3. Plot of weight loss against time for Al corrosion in 1M HCl with JCLE at 50°C
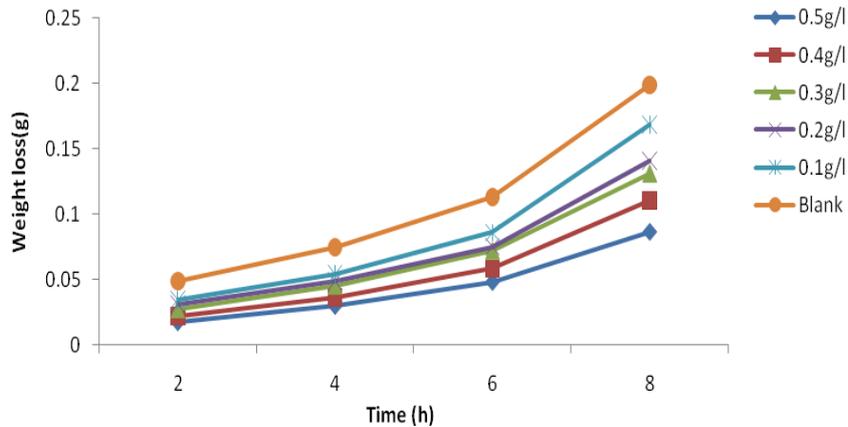

Figure 4. Plot of weight loss against time for Al corrosion in 1M HCl with JCLE at 60°C

The Inhibition efficiency, Є (%) was evaluated using Eqn. 2 [28].

$$\text{Є (\%)} = [W_{blank} - W_{inh} / W_{blank}] \times 100 \qquad (2)$$

where W_{blank} and W_{inh} are the corrosion rates in the absence and presence of the inhibitor respectively

Figures 1-4 show the weight loss against time for aluminium corrosion with and without different concentrations of the JCLE at 30°C - 60°C.

A close observation of the figures show that weight loss of aluminium in HCl varied with time and was reduced in the presence of JCLE compared to the blank.

Weight loss also increased with rise in temperature, with the highest weight loss attained at 60°C. The increase in

weight loss with temperature is an indication that inhibition efficiency decreased with increasing temperature suggesting possible desorption of some of the adsorbed JCLE molecules from the metal surface at higher temperatures. From Fig. 5, the inhibition efficiencies of 0.5g/L of JCLE extract were 76.49, 71.95, 63.07 and 56.55% respectively at 30, 40, 50, and 60°C. It can be inferred from this trend that the JCLE was physically adsorbed on the metal surface. This trend may be due to the fact that most effects at elevated temperatures are adverse to corrosion inhibition by increasing the corrosion rate and decreasing the tendency of the JCLE to be adsorbed on the aluminium surface. Similar results have been reported by other researchers [3, 9]. Inhibition efficiency (Fig. 5) increased with JCLE concentration due to an increase in the metal surface area covered by the extract [7], with the highest inhibition efficiency obtained with 0.5g/L.

Surface area coverage plays an important role in fitting experimental data to adsorption isotherms which consequently helps in determining the inhibition mechanism. Figure 6 shows an increase in corrosion rate of the metal due to temperature increase; however increase in the concentration of the extract lead to a remarkable decrease in the corrosion rate. This remarkable decrease in weight loss/corrosion rate is as a result of increased inhibition efficiency with increasing JCLE concentration indicating that more inhibitor molecules are adsorbed on the aluminium surface leading to the formation of a protective film. Similar results for the inhibition of corrosion of Al in 2M HCl by exudate gum from *Dacroydes edulis* has been reported [31].

Adsorption Consideration: The degree of surface coverage values from weight loss measurement has a direct relationship with inhibition efficiency given by $\theta = \%\ \epsilon\ /100$ [29]. This surface coverage is a direct consequence of the phytochemicals contained in the JCLE (Table 1) which have various degrees of heteroatoms that interact with the metal surface [8]. Adsorption of inhibitor involves the formation of two types of interaction; physisorption and chemisorption.
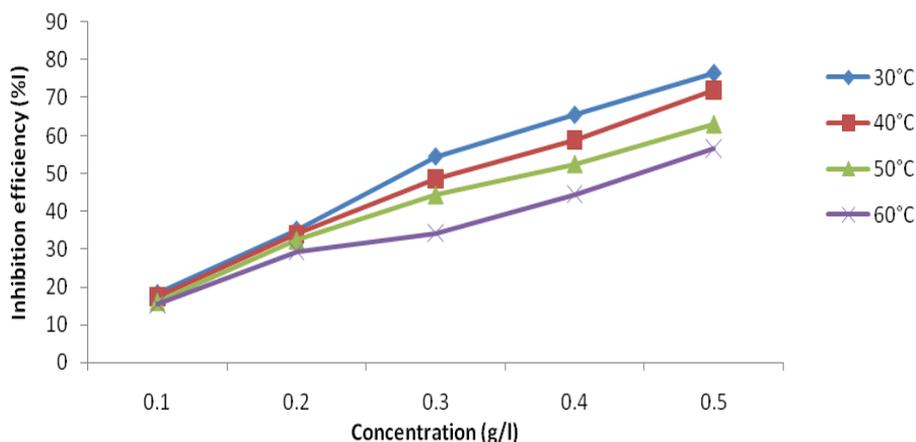

Figure 5. Plot of inhibition efficiency vs. concentration for Al corrosion in 1M HCl with JCLE
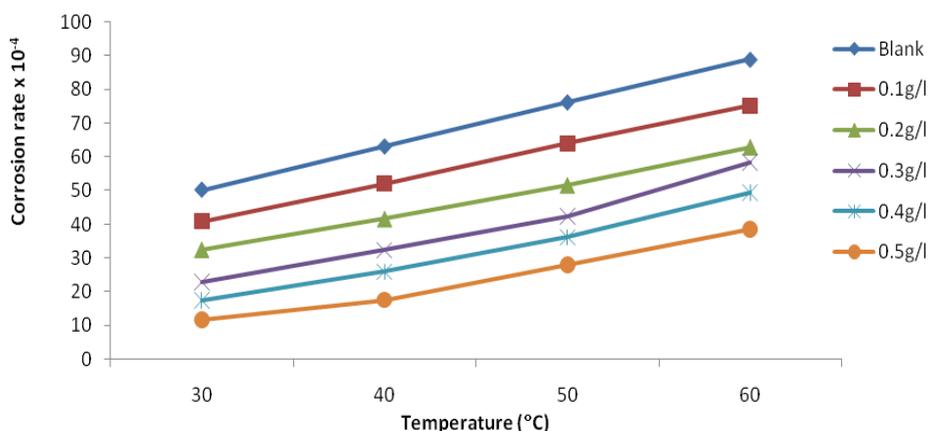
Figure 6. Plot of corrosion rate against temperature for Al corrosion in 1M HCl with JCLE

Table 1. Phytochemicals of Jatropha curcas leave extract*

Phytochemicals	Tannins	Terpenes	Anthra- quinone	Alkaloids	Flavonoid	Deoxy sugar	Cardiac glycoside	Saponins
Presence	+++	+++	+++	+++	+++	++	++	+

* +++ = highly present; ++ = moderately present; + = present in trace amount

To ascertain the degree of adsorption, the surface coverage for JCLE in HCl and on aluminium corrosion for 30°C, 40°C, 50°C, 60°C were fitted into different adsorption isotherm model and correlation coefficient (R^2) value were used to determine best fit.

Freundlich adsorption isotherm was tested for compliance to the data obtained during the experiment using Eqn. 3.

$$\theta = kC^n \text{ where } 0 < n < I \qquad (3)$$

Eqn. 3 could be written in the linear form is written as Eqn. 4.

$$In\ \theta = Ink + n\ In\ C \qquad (4)$$

where k and C, represents the equilibrium constant of adsorption and extract concentrations respectively.

A plot of In θ against In C gave a straight line as applicable by Eqn. 4 which suggest that the experimental data obeyed Freundlich adsorption isotherm providing evidence for the role of adsorption in the observed inhibitive effect of the extract. Temkin adsorption isotherm was tested using Eqn. 5.

$$KC = \exp(2a\theta) \qquad (5)$$

where a is the molecules interaction parameter, θ is the degree of surface coverage of the inhibitor on the Al, K is the equilibrium constant of adsorption, and C is the extract concentration.

A plot of θ against long C was linear (Fig. 8), suggesting that the experimental data fits Temkin adsorption isotherm.

El-Awady adsorption isotherm was also tested. The characteristics of the isotherm are given by Eqn. 6.

$$Log[\theta/1-\theta]= log\ K + y\ log\ C \qquad (6)$$

where C is the concentration of the extract, θ is the degree of surface coverage, K_{ad} is the equilibrium constant of adsorption process which is equals to $K^{1/y}$.

In this model, the number of active site y is included, values of 1/y <1 suggest multilayer adsorption while 1/y > 1 suggest that a given inhibitor molecule occupied more than one active site [9]. A plot of log [θ/1-θ] against log C was also linear with the highest correlation of 0.9935. The values of 1/y and k_{ad} calculated from the E1-Awady et al model curve is given in Table 2.

y = 0.8847x + 0.2895
R² = 0.9968

y = 0.9072x + 0.4036
R² = 0.9935

y = 0.8398x + 0.1473
R² = 0.9913

y = 0.7804x - 0.0702
R² = 0.9844
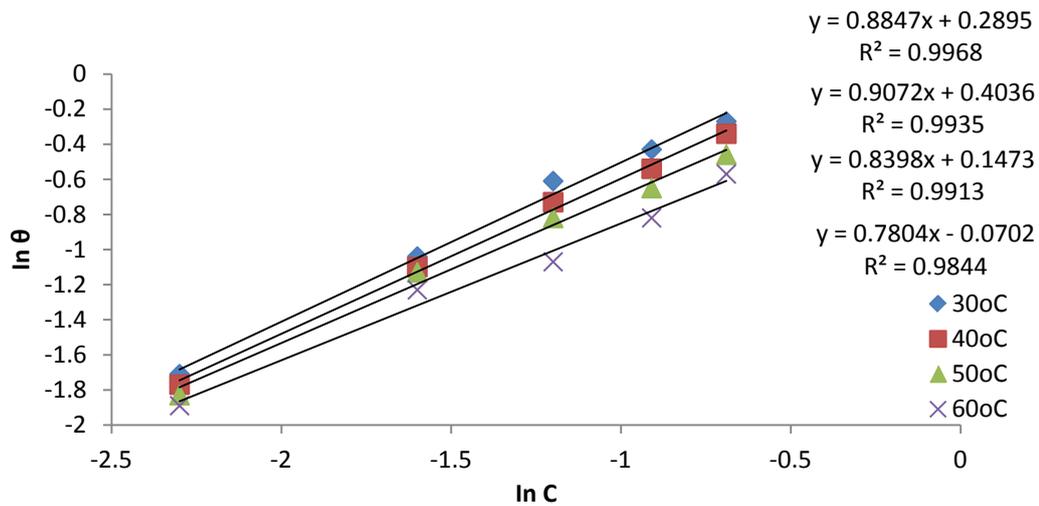
Figure 7.   Freundlich adsorption isotherm for Al corrosion in 1M HCl with JCLE at 30-60°C

y = 0.6497x + 0.7908
R² = 0.9811

y = 0.8304x + 0.9776
R² = 0.9764

y = 0.7505x + 0.8893
R² = 0.9695

y = 0.5411x + 0.6698
R² = 0.9378
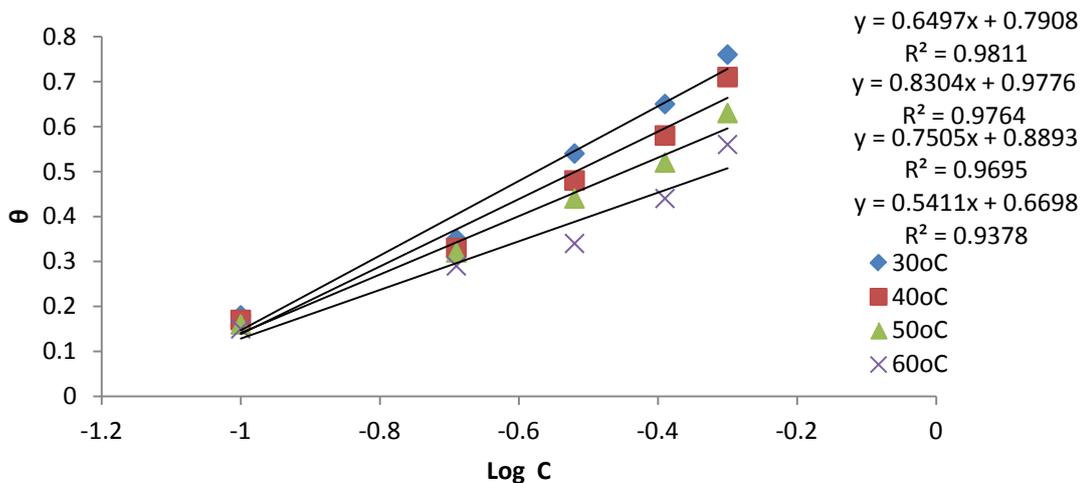
Figure 8.   Temkin adsorption isotherm for Al corrosion in 1M HCl with JCLE at 30 - 60°C

$y = 1.635x + 0.9223$
$R^2 = 0.9879$

$y = 1.4746x + 0.7532$
$R^2 = 0.9858$

$y = 1.3102x + 0.5839$
$R^2 = 0.9935$

$y = 1.1536x + 0.3811$
$R^2 = 0.9736$
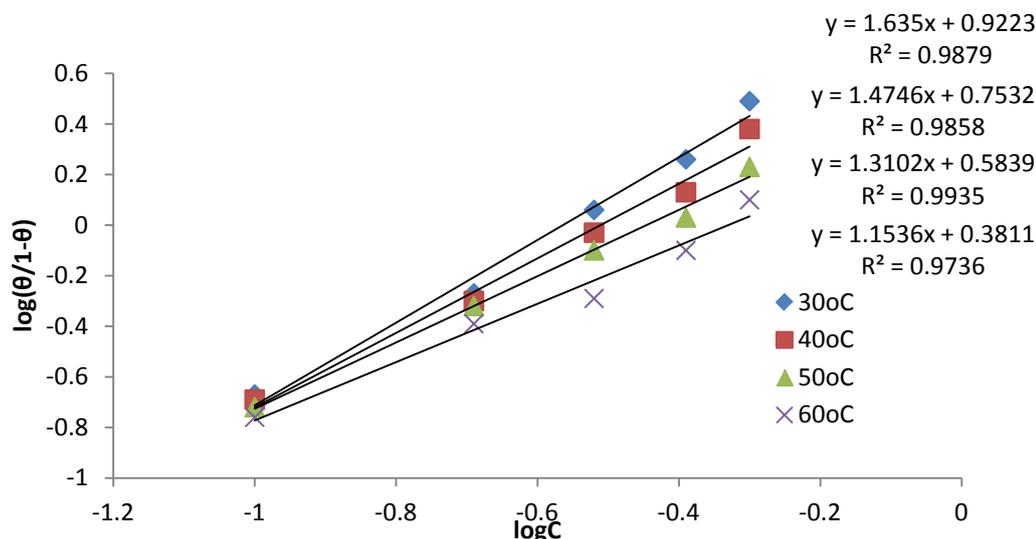
Figure 9. El-Awady adsorption isotherm for Al corrosion in 1M HCl with JCLE at 30-60°C

Effect of temperature:

The effect of temperature on inhibition reaction is highly complex, occasioned by many changes that may occur on the metal surface such as etching, rupture and desorption of inhibitor. Thus temperature can modify the interaction between the mild steel and the solution medium [8]. On a generally note, the rate of reaction (corrosion) increases with rise of temperature. The standard free energy of adsorption is associated with water adsorption/desorption equilibrium which forms an important part in the overall free change of adsorption. The equilibrium constant for adsorption process from El-Awady et al. adsorption isotherm (K) is related to the standard free energy of adsorption by Eqn. 6.

$$K = 1/55.5 \exp(-G_{ads}/RT) \qquad (6)$$

where, T is the absolute temperature; R is the molar gas constant (8.314 kJ/mol); 55.5 is the concentration of water in solution expressed in mol/L.

The negative values of ΔG_{ads} (Table 2) clearly confirmed the spontaneity of the adsorption process and consequently the stability of the strongly adsorbed layer on the aluminium surface [32, 33]. Values of ΔG_{ads} up to -20 kJmol^{-1}, like the ones obtained with JCLE, are consistent with electrostatic interaction between the charged metal, which indicates physical adsorption, while those more negative than -40 kJmol^{-1} involves charge sharing or transfer from the inhibitor molecule to the metal surface to form a co-ordinate type of bond (which indicates chemical adsorption) [31].

Table 2. Adsorption parameters for the adsorption of JCLE in 1M HCl on Al at different temperatures

Temp (°C)	K_{ads}	ΔG_{ads}(kJ/mol)	1/y	R^2
30	8.356	-16.99	0.61	0.987
40	5.662	-14.98	0.67	0.985
50	3.828	-14.38	0.76	0.993
60	2.404	-13.54	0.86	0.973

The values of ΔG_{ads} obtained in our study are between -13.54 and -16.99 kJ mol^{-1} suggesting that the type of adsorption involved in JCLE attachment to aluminium surface is physiosorption. The values of ΔG_{ads} given in Table 2 indicate that JCLE are adsorbed on the aluminium surface through physisorption. The ΔG_{ads} reduces negatively with increase in temperature indicating that the adsorption is unfavourable with increasing experimental temperature [34]. It could be concluded that the variation %ε with temperature (Fig. 5) and higher values of Ea and Q_{ads} (Table 3) in the presence of additives compared to blank indicates major contribution of physiosorption in the adsorption process [8].

In order to elucidate the inhibitive properties of the JCLE and the temperature dependence on the corrosion rates, the apparent activation energy (Ea) for the corrosion process without and with different concentrations of the inhibitor were calculated from Arrhenius type plot according to Eqn. 7 [35]. The results are presented in Table 3

$$\text{Log}(v_2/v_1) = (Ea/2.303R)(T_2 - T_1/T_1 x T_2) \qquad (7)$$

Where v_1 and v_2 are the corrosion rates at absolute temperatures T_1 and T_2, respectively and R is a molar gas constant.

A change in the value of Ea in presence of JCLE may be due to the modification of the mechanism of the corrosion process in presence of adsorbed inhibitor molecules. Bouyanzer et al., [36] indicated that generally, higher values of Ea in presence of additives support physical adsorption mechanism whereas an unchanged or lower value of Ea for inhibited systems compared to the blank is indicative of chemisorption mechanism. The inhibitor causing an increase in the values of Ea compared to blank retard the corrosion at ordinary temperature, but the inhibition is diminished at elevated temperature. From Table 3, it is clear that Ea for the corrosion process in the presence of JCLE extracts are higher than those in the absence of the extracts (blank). The higher activation energies mean a slower

reaction and that the reaction rate is very sensitive to temperature.This observation is consistent with physical adsorption mechanism which leads to the formation of an adsorptive film of electrostatic character [8].

An estimate of the heat of adsorption (Q_{ads}) was obtained from the trend of surface coverage (θ) with temperature according to Eqn. 8 [37]. Temperature of 30°C was taken as T_1 while the highest temperature under study which was 60℃ was taken as T_2.

$$Q_{ads}= 2.303R [\log (\theta_2/1-\theta_2) - \log (\theta_1/1-\theta_1)]$$

$$x (T_1T_2/T_2-T_1) \text{ kJ mol}^{-1} \qquad (8)$$

where θ_1 = degree of surface coverage at temperature T_1, θ_2 = degree of surface coverage at temperature T_2.

Negative Q_{ads} values are consistent with physisorption characteristics with inhibition efficiency decreasing with increase in temperature while positive values of Q_{ads} suggest increased inhibition efficiency at higher temperature. The calculated values of Q_{ads} (Table 3) for aluminium in the acid with JCLE were all negative and were of the range of -6.11 to -25.33kJ/mol which is consistent with the proposed inhibition physisorption characteristics [37].

Table 3. Calculated values of rate constant (k), activation energy (Ea), heat of adsorption (Q_{ads}) for aluminium corrosion in 1M HCl with

Conc. (g/L) of JCLE	Ea (kJ mol^{-1})	Q_{ads} (kJ mol^{-1})
0.0	15.52	-
0.1	16.67	-6.11
0.2	17.99	-7.72
0.3	25.62	-22.98
0.4	28.51	-24.02
0.5	33.09	-25.33

At all the temperatures studied, the adsorption process of the leave extract on the aluminium coupon surface best obeyed Freundlich adsorption isotherm (Table 4) and least obeyed the Temkin adsorption isotherm.

Table 4. Comparison of the isotherm R^2

Temp(°C) / Isotherm type	Freundlich	El -Awady	Temkin
30	0.9968	0.9878	0.9811
40	0.9935	0.9858	0.9764
50	0.9913	0.9935	0.9695
60	0.9844	0.9736	0.9378

Freundlich adsorption isotherm has been reported [17, 37] to have best described experimental data obtained by the inhibitory actions of plant extracts in acidic medium and corrosion inhibition of aluminium in 2M HCl and 2M KOH by *Sansevieria trifasciata* extract respectively. Umoren et al [31] though reported Temkin adsorption isotherm to have satisfactorily explained data obtained from corrosion inhibition of exudates gum from *Dacroydes edulis* on aluminium metals in hydrochloric acid medium.

4. Conclusions

Jatropha curcas leave extract have been found to act as corrosion inhibitor on aluminium in 1M hydrochloric acid solution. The maximum inhibiion efficiency of the extract was found to be 76.49% at a concentration of 0.5g/L of the extract. The adsorption of JCLE on the surface of aluminium in 1M HCl followed Freundlich, El-Awady and Temkin adsorption isotherms, in that order. The calculated values of Q_{ads} for aluminium in the acid with JCLE were all negative and were of the range of -6.11 to -25.33kJ/mol. A mechanism of physical adsorption is proposed for the process.

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