

# Corrosion Inhibitor Activity of Extract of *Jatropha Curcas* Leaves for Mild Steel in HCl Medium

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## Abstract

*Jatropha curcas* leaves extract was investigated as a green inhibitor on the corrosion of mild steel in aqueous hydrochloric acid solution using gravimetric and thermometric techniques. The inhibition efficiency increased with increase in concentration of the leaves extract. Maximum inhibition efficiency was found to be 95.92% in 2M HCl with 10ml concentration of the extract in gravimetric method. In thermometric method, the maximum inhibition efficiency was found to be 87.04% at the same concentration. The inhibiting effect was attributed to the presence of alkaloids, flavonoids, saponins, tannins and phenol in the extract. It was found that *Jatropha curcas* leaves extract obeys Langmuir adsorption isotherm. From the results, a physical adsorption mechanism is proposed for the adsorption of *Jatropha curcas* leaves extract on mild steel surface.

**Keywords:** *Jatropha curcas*, leaf extract, corrosion, Langmuir isotherm, adsorption, steel.

## Introduction

The word corrode is derived from the Latin word *corrode*, meaning to eat into or wear gradually. Corrosion is a naturally occurring process generally defined as the deterioration of a material and/or its properties because of reaction with its environment [1]. Corrosion is a naturally occurring phenomenon which affects our society daily, and it results into damage, destruction and degradation to household gadgets, automobiles, airplanes, highway bridges, energy production and distribution systems, among others [2].

Corrosion is one of the major serious global problems in industries and resulting to losses each year in hundreds of billions of dollars. Corrosion studies have been carried out in several countries and their finding showed that annual corrosion costs ranged

approximately from 1 to 5% of the Gross National Product (GNP) of each nation [3]. The consequences of corrosion are many, and the effect of these on safe, reliable and efficient operation of equipment or structure is often more serious than simple loss of a metal. Some of the major harmful effects of corrosion include reduction of metal thickness, hazards or injuries to people arising from structural failure, loss of time, reduced value of goods, contamination of fluids in vessels and pipes, etc. [4].

Mild steel is a material of choice and it finds wide application in many industries like automobile, petrochemicals, construction, metallurgical, refineries, and so on. This is due to its low cost, availability and excellent mechanical properties [5]. However, it suffers corrosion in hostile aggressive acidic or alkaline environment. Acids are used to remove oxides and other contaminants from metal surfaces. Acids are also used for derusting and pickling, the cleaning of refinery equipment oil well acidizing, descaling, removal of calcareous deposits from boilers, radiators from vehicles, pipelines carrying petroleum products, etc. [6].

There are numerous various forms of controlling the corrosion of metals such as material selection, coatings, cathodic protection, design and use of inhibitors. However, the use of inhibitors is one of the best methods of protecting metals against corrosion [4]. An inhibitor is a chemical that when added to corrosive environment in small concentration reduce the rate of metal corrosion [7]. These inhibitors can be anodic, cathodic, mixed or volatile corrosion inhibitors [8]. Most of the corrosion inhibitors are synthetic chemicals, which are expensive and very hazardous to environment. Therefore, it is desirable to source for environmentally safe and green inhibitors, which are non-toxic, biodegradable and readily available [9].

Several researchers have reported the use of plant extracts as effective corrosion inhibitor for mild steel in acidic media. These plant extracts include *Rosemarinus Officinalis* [10], *Parica Papaya* [11], *Azadirachta Indica* [12], *Vernonia Amydalina* [13], *Carica Papaya* [14], *Allium Sativum* [15], *Phyllanthus Amarus* [16], *Telfaria Occidentalis* [17], *Sansevieria Trifasciata* [18], *Hibiscus Subdariffa* [19], *Lawsonia* [20], *Musa Sapientum* [21], *Prosopis Cineraria* [22], *Gossipium Hirsutum* [23] among others. In a recent study, *Jatropha Curcas* leaves extract was found to act as a good corrosion inhibitor for mild steel in aqueous sulphuric acid solution. The inhibition action was dependent on the concentration of the leaves extract in the acid solution [24]. This study is aimed at investigating the

effectiveness of the leaves extract of *Jatropha Curcas* as corrosion inhibitor of mild steel in hydrochloric acid medium using gravimetric and thermometric methods.

## Materials and methods

The mild steel sheet was mechanically pressed cut into 4cm X 2cm and 1.5cm X 2cm coupons. The specimens were polished using LINN MAJOR STRUER-ITALY (Model No. 224732) with emery papers 140/0304 – 140/0308 grades. Subsequently, they were degreased in ethanol, dried in acetone and stored in a desiccator. The chemical composition of the mild steel sample used for this experiment was analyzed using optical emission spectrometer and the results are presented in Table 1.

Table 1 Chemical composition of the mild steel sample

S/N	Element	wt.%
1.	Carbon(C)	0.17
2.	Silicon(S)	0.21
3.	Manganese(Mn)	0.55
4.	Phosphorus(P)	0.02
5.	Sulphur(S)	0.02
6.	Copper(Cu )	0.18
7.	Nickel(N)	0.01
8.	Tin(Sn)	0.02
9.	Iron(Fe)	98.81

The fresh leaves of *Jatropha Curcas* (JC) plant was obtained, washed under running water, cut into pieces, air dried and then grounded well and sieves into powdery form. Then, 10g each of the powdery leaf was put into flat bottom flask containing 200cm<sup>3</sup> of 2M HCl solution. The resulting solution was refluxed for 2 hours and left overnight before it was

carefully filtered. The stock solution was prepared from the filtrate and prepared into the desired concentrations. The experimental set up and procedure followed the method adopted by Rosaline et al. [25]. Pre-treated mild steel specimens of size 1.5cm X 2cm X 0.2cm were weighed prior to immersion into HCl solution using an electronic balance. The weighed samples of mild steel were immersed completely in 50ml of 2M HCl for 5 days. The mild steel specimens were taken out of the solution every 24 hours, washed thoroughly with distilled water, and dried completely. The final weights were noted. From the initial and final weights of the specimen for each day, the loss in weight was calculated.

From the weight loss, the corrosion rate, inhibition efficiency (IE) % and surface coverage ( $\theta$ ) of the plant extract were calculated using the Equations 1, 2 and 3 [26, 27]:

$$\text{Corrosion rate (gcm}^{-2} \text{ h}^{-1}) = \frac{\Delta W}{AT} \quad (1)$$

$$\text{Inhibition Efficiency (IE \%)} = \left( \frac{CR_B - CR_W}{CR_B} \right) \times 100 \quad (2)$$

$$\text{Surface Coverage } (\theta) = \left( \frac{CR_B - CR_W}{CR_B} \right) \quad (3)$$

where  $CR_B$  and  $CR_W$  are corrosion rates in the absence and presence of the inhibitor,  $W$  is the weight loss (g),  $A$  is the surface area of the mild steel coupon ( $\text{cm}^2$ ) and  $T$  is the time of exposure (hours).

Thermometric analysis was carried out according to the method described by Ejikeme et al. [28]. The mild steel coupons of dimension 4cm X 2cm X 0.2cm were immersed in 50 ml of 4M HCl solution as shown in Figure 1. The initial temperature was recorded. The process of the corrosion reaction was monitored by determining the change in temperature with time using a standard digital thermometer.



Figure 1. Thermometric experimental set up

From the rise in temperature per minute, the reaction numbers (RN), inhibition efficiency (IE) and surface coverage ( $\theta$ ) were calculated using Equations 4, 5 and 6 respectively:

$$RN (^{\circ}C \text{ min}^{-1}) = \frac{T_m - T_i}{t} \quad (4)$$

$$\%IE = \left( \frac{RN_{aq} - RN_{wi}}{RN_{aq}} \right) \times 100 \quad (5)$$

$$\theta = \left( \frac{RN_{aq} - RN_{wi}}{RN_{aq}} \right) \quad (6)$$

where RN = Reaction Number

$RN_{aq}$  = Reaction Number in the absence of inhibitor (Control)

$RN_{wi}$  = Reaction Number in the presence of inhibitor

$T_m$  = Maximum Temperature attained by the system

$T_i$  = initial temperature

$T$  = Time (minutes)

## Results and discussion

The phytochemical composition of *Jatropha curcas* leaves is shown in Table 2. The result showed that the extract contains tannins, saponins, alkaloids, phenol and flavonoid. It has been reported that saponins, tannins and alkaloids are active constituents of most green inhibitors [29].

Table 2 Results of major phytochemical screening of the constituents of *Jatropha Curcas* leaves extract

Phytochemical	Alkaloid	Flavonoid	Saponins	Tannins	Phenol
Composition(mg/L)	1.610	0.672	0.412	0.124	0.465

The curves for the variation of weight loss with exposure time for the mild steel specimens immersed in 2M HCl solution with varied concentrations of *Jatropha Curcas* leaves extract are presented in Figure 2. From the figure, it was found that the weight loss of the mild steel specimens increased with the period of exposure but decreased as the concentration of the inhibitor increases. This indicates that the rate of corrosion of mild steel increases with increase in the period of exposure, and that *Jatropha Curcas* leaves extract inhibited the corrosion of mild steel in 2M HCl. The order of increasing corrosion inhibition performance was: 10ml>8ml>6ml>4ml>2ml concentration of the extract of *Jatropha Curcas* leaves. Undoubtedly, the constituents of *Jatropha Curcas* leaves extract exhibited a reasonable degree of electrochemical corrosion inhibition activity that was dependent on the concentration. Loto [30] reported that the corrosion inhibition performance could be attributed to the presence of alkaloids and synergistic combination of other phytochemical constituents which form a barrier on the surface of mild steel as observed when *Camellia Sinensis* was used as an extract for the inhibition of the corrosion of mild steel in dilute sulphuric acid.



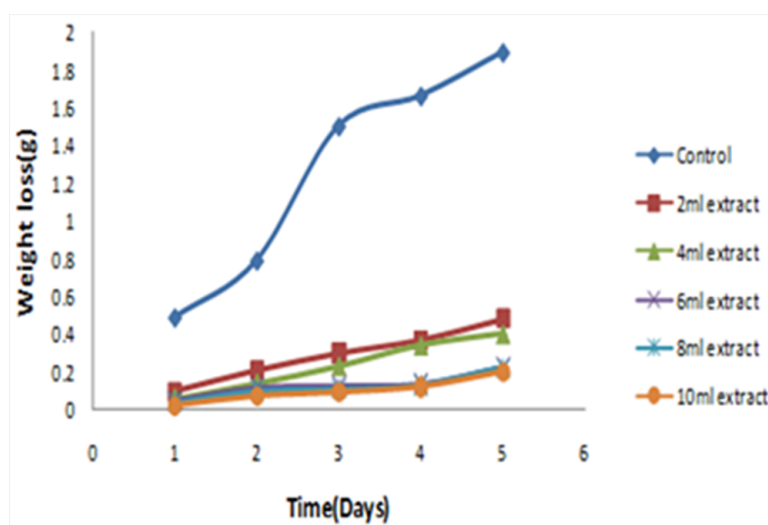


Figure 2. Variation of weight loss with time for the corrosion of mild steel in 2M HCl.

Table 3 shows the result of inhibition efficiencies of *Jatropha Curcas* leaves extract on corrosion of mild steel specimens in 2M HCl at different time of immersion. From the table, it is observed that inhibition efficiency increases as the concentration of the extract of JC increases at all the exposure times. At 120 hours of exposure, the inhibition efficiency was 74.60% for 2ml extract concentration while for 10ml concentration, the inhibition efficiency was 89.42%. The inhibition efficiency of the leaves of *Jatropha Curcas* extract could be associated with its complex chemical compound which include alkaloid as reported earlier [29]. The phytochemical constituents act as inhibitive passive film formers on the mild steel. Umoren et al. [31] reported similar experimental results to the present work when *Aningeria robusta* extract was used as green inhibitor in HCl solution and observed that as the concentration of the inhibitor increases, the inhibition efficiency increases. They also reported that the adherent film of the inhibitor on the surface of immersed specimens hindered active corrosion reactions, and hence the penetration of the  $\text{Cl}^-$  reacting species through film barrier. The synergistic action of the constituents promote more stable passive film formation on the surface of the mild steel and thereby increasing the inhibition efficiency of the plant extract as reported by Patel et al. [32].

Table 3 Inhibition efficiency of mild steel in different concentrations of Jatropha Curcas leaves extract in 2M HCl at different time of immersion.

Time (Days)	Inhibition efficiency (I.E. %) of J.C. leaves extract in HCl				
	2ml	4ml	6ml	8ml	10ml
1	79.59	87.76	89.8	91.84	95.92
2	73.42	78.48	84.81	87.34	91.14
3	80.00	84.67	91.33	92.67	94.00
4	77.71	79.52	91.57	92.17	92.77
5	74.60	78.84	87.83	88.36	89.42

Figure 3 shows the variation in corrosion rate with time of exposure with varying concentration of Jatropha Curcas leaves extract. The result obtained showed that the rate of corrosion increased from 0 g/cm<sup>2</sup> at the beginning of the experiment to a value of  $6.80 \times 10^{-3}$  g/cm<sup>2</sup>hr after 24 hours of exposure due to initial corrosion attack. Subsequently, the corrosion rate decreased to a value of 5.49 g/cm<sup>2</sup>hr after 48 hours exposure. This may be due to the formation of corrosion products, which tend to shield the metal surface from corrosion attack, thereby reducing the rate of corrosion as reported by Nnanna et al. [33]. Above 48 hours exposure, the corrosion rate increases up to a value of 6.94 g/cm<sup>2</sup>hr probably due to breaking down of the protective bond of the inhibitor leading to increased corrosion rate. From 72 to 120 hours immersion period, the rate of corrosion decreases, with the corrosion rate at 5.25 g/cm<sup>2</sup>hr after 120 hours. For 2ml inhibitor concentration, the corrosion rate increases from 1.40 g/cm<sup>2</sup>hr at 24 hours exposure time to 1.46 g/cm<sup>2</sup>hr after 48 hours exposure which could possibly be due to surface corrosion attack. However, above 48 hours of exposure, the corrosion rate decreases to a value of 1.33 g/cm<sup>2</sup>hr after 120 hours exposure. This decrease in corrosion rate can be attributed to the adsorption of the molecules of the inhibitor on the surface of mild steel [34]. Amitha and Bharathi [35] reported that that green inhibitors contain phytochemical constituents which act as physical barrier that restrict the diffusion of ions and then prevent metals from corrosion attack. However, at 4ml inhibitor concentration, the corrosion rate further increase with time possibly due to the breaking down of the protective film [29]. For 6–8ml inhibitor



concentrations, it was observed that the corrosion rate increased gradually up to an exposure time of 72 hours and later decreased for all other exposure times.

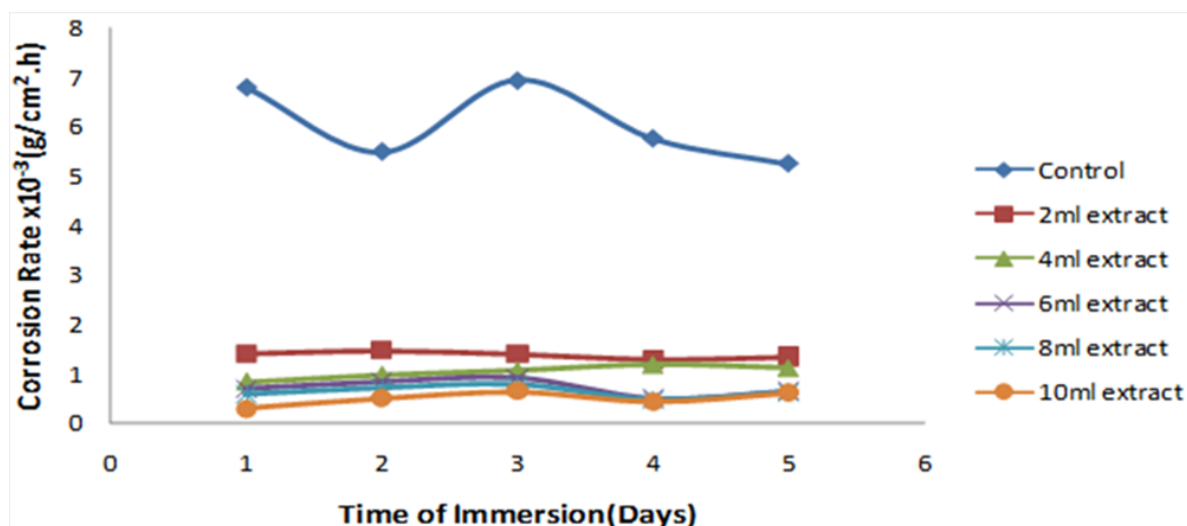


Figure 3. Variation in corrosion rate against time of immersion of the extract in 2M HCl.

Figure 4 shows the variation of volume of hydrogen gas evolved with time for the corrosion of mild steel in various concentrations of the inhibitor. The figure revealed that the volume of hydrogen gas evolved increased with increasing period of exposure, but decreases with increase in the concentration of acidic extract of *Jatropha Curcas* leaves. The volume of hydrogen gas evolved at 30 minutes was 21.8 cm<sup>3</sup> for the blank solution, while that of 4ml, 6ml, 8ml and 10ml concentrations of *Jatropha Curcas* leaves extract are 9.0, 8.0, 6.8 and 5.0 cm<sup>3</sup>, respectively. The blank system having no inhibitor gave the highest hydrogen gas evolution and is far apart when compared to when varying concentration of the extract of *Jatropha Curcas* leaves was added. This may be due to the absence of inhibitor that will prevent acidic solution from reaching the metal surface as reported by Al -Turkustani et al. [36]. It was further reported by the authors that presence of oxide film causes the rate of hydrogen gas evolution to decrease (i.e. decrease in the rate of corrosion). This agrees with the observation of Aisha [37], which reported that increase in hydrogen evolution gas in the blank system may be due to direct reaction of the acid with the surface of mild steel, since there is no adsorption layer to inhibit the reaction of the acidic media on the surface of the sample. Hence, the rate of hydrogen gas evolution, that is, the corrosion rate will be faster in the blank solution (without inhibitor) compared with the inhibited acidic solution. Ulaeto et al. [38] found that the leaf and root extracts of *Eichornia Crassipe* effectively inhibited the corrosion of mild steel in 5M HCl, and that the extracts performed better at higher concentrations.

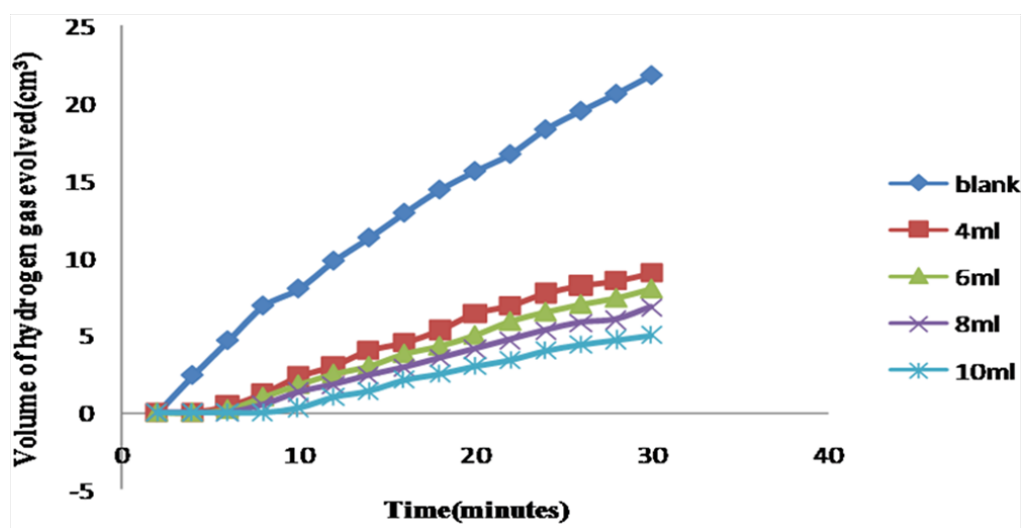


Figure 4. Variation of volume of  $H_2$  ( $cm^3$ ) evolved with time (minutes) of mild steel coupons for different volumes of JC extract in HCl solution

The variation of inhibition efficiency against time of immersion with varying concentration of the inhibitor is shown in Figure 5. The results show that from 0 to 4 minutes, the inhibition efficiency was 0%. Above 4 minutes, the corrosion rate became faster resulting in higher inhibition efficiency. However, at 6 minutes there was a re-ordering of the inhibition efficiencies from highest to the least value in descending order of the inhibitor concentration i.e. ( $10ml < 8ml < 6ml < 4ml$ ) at all the exposure time. This revealed that there is an adsorption of the constituents of the *Jatropha Curcas* leaves extract on the surface of mild steel with 10ml concentration of the inhibitor having the highest inhibition efficiency. It was reported by Kuznetsov [39] that the longer the latency period, the higher the inhibition efficiency. In addition, the chemical composition of the plant extracts also retard metal dissolution [39].

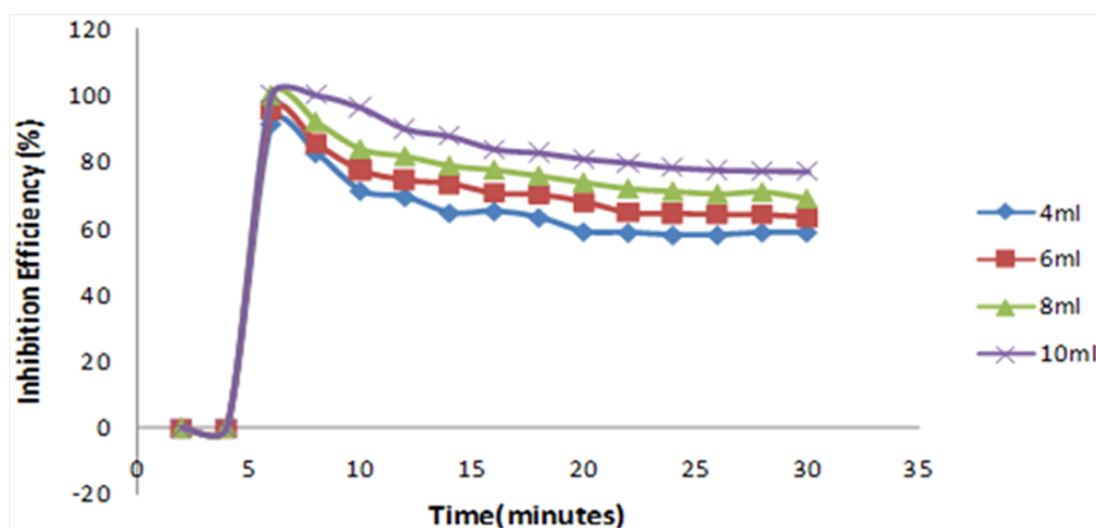


Figure 5. Variation of inhibition efficiency with the time of immersion in HCl

Figure 6 reveals the results obtained from the variation of the temperature with time of immersion for mild steel corrosion in 4M HCl with and without extract of *Jatropha curcas* leaves at different concentrations of the inhibitor. The results revealed that maximum temperature was attained in the blank solution. This corresponds to a reaction number of 0.633°C/min (Table 4). Further inspection of Figure 6 revealed that on addition of the extract, the maximum temperature decreased. Similar results have been reported that maximum temperature decreased when various concentrations of plants extract were used, and this has been attributed to the formation of a barrier layer when the extract adsorbed on the surface of the metal [6, 28, 40]. This is in agreement with the current study. The calculated values of reaction number (RN), percentage reduction in reaction number (inhibition efficiency) and surface coverage for various concentrations of *Jatropha Curcas* extract is presented in Table 4. The result showed that the reaction number which corresponds to the rate of corrosion of mild steel decreased in the presence of *J. Curcas* leaves extract compared to the blank solution. In addition, the percentage inhibition and the degree of surface coverage increase with increase in *J. Curcas* extract concentration. Similar observations have been reported by several authors, which supported the results in the present study [38, 39].

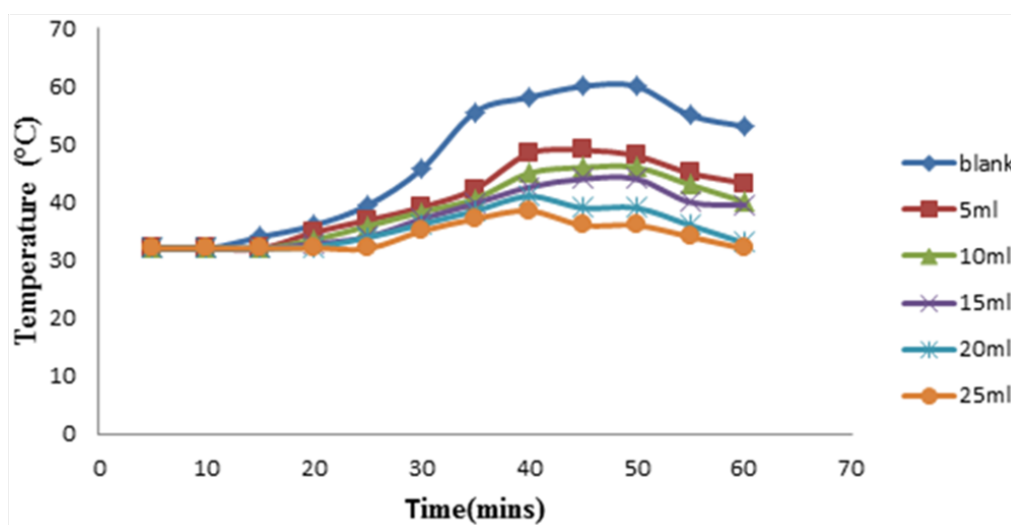


Figure 6. Temperature–time curves for mild steel corrosion in 4M HCl with and without extract of *Jatropha Curcas* leaf at different concentrations of the inhibitor

Table 4 Calculated values of Reaction number, percentage inhibition efficiency and the degree of surface coverage at different concentration of *J. Curcas* leaf extract in 4M HCl

Concentration of JC extract (ml)	Reaction Number (°C/min)	Inhibition Efficiency (%I.E.)	Surface Coverage ( $\theta$ )
Blank	0.633	–	–
5.00	0.387	38.8	0.3880
10.00	0.316	50.08	0.5008
15.00	0.269	57.50	0.5750
20.00	0.164	74.09	0.7409
25.00	0.082	87.04	0.8704

Figure 7 shows the variation of  $\log C/\theta$  against  $\log C$  for the extract of *Jatropha curcas* in HCl aqueous solution. The results show a clear linear relationship with a regression close to unity of 0.999 for *Jatropha Curcas* leaf extract in HCl. This proves that Langmuir isotherm model is appropriate for the adsorption of the extract of *Jatropha Curcas* leaves adsorption

in HCl. Plant extract contains organic compounds having polar atoms or groups which are absorbed on the metal surface. Obot and Obi-Egbedi [40] reported that compounds interact by mutual repulsion or attraction and this may be advocated as the reason for the departure of the slopes values from unity when *Ipomoea Involcrata* was used as plant extract. The application of Langmuir isotherms to the adsorption of *Jatropha Curcas* leaves extract on mild steel indicated that there is no interaction between the adsorbate and the adsorbent. The application of Langmuir isotherm to some plants extract on the surface of metal suggests that there is no interaction between the adsorbed species as reported by some authors [26, 27].

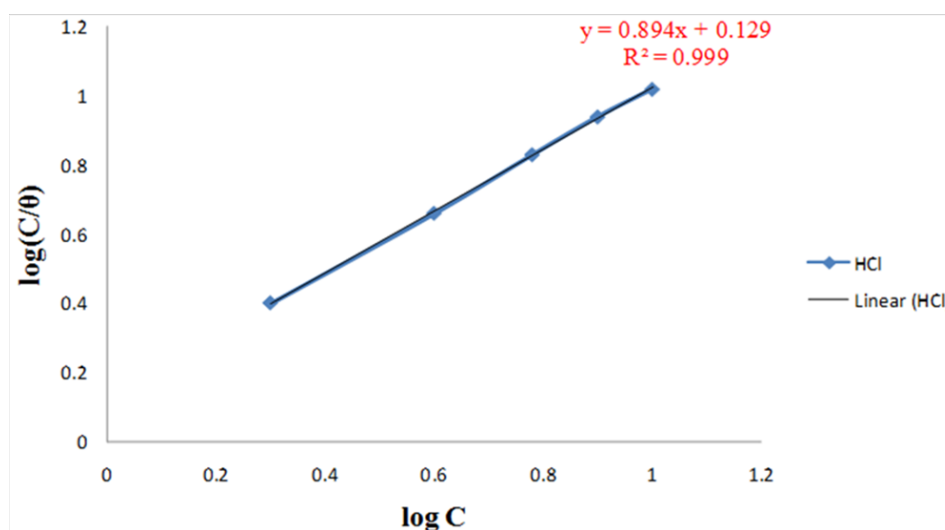


Figure 7. Langmuir adsorption isotherm plot of  $C/\theta$  against  $C$  for HCl

The values of Langmuir adsorption isotherm parameters of *Jatropha Curcas* leaves extract in 2M HCl is given in Table 5. As shown in the table, the calculated values of the standard free energy of adsorption have negative values. This indicates the spontaneity of the process. The value of the standard free energy of  $-10.9$  kJ/mole shows that the adsorption is within an order of  $-20$  kJ/mol. This measured value of  $\Delta G_{ads}$  suggests a strong physical adsorption of green inhibitors leaves extract onto the surface of mild steel in 2M HCl solution.

Table 5 Calculated values of Langmuir Adsorption isotherm parameters of *Jatropha Curcas* Extract in HCl

Plant Extract	Intercept	Slope	Log K	R <sup>2</sup>	$\Delta G_{ads}$ (KJ/mol)
JC in 2M HCl	0.668	0.894	0.129	0.999	-10.9

The plots of  $\log (\theta / C)$  versus  $\log (1-\theta)$  as shown in Figure 8 does not give a perfect fit, indicating that application of Flory–Huggins isotherm to the adsorption of extract of leaves of *Jatropha curcas* on the surface of mild steel is not suitable. The values of the adsorption parameters deduced from Flory–Huggins plots are presented in Table 6. The correlation coefficients ( $R^2$ ) were gotten from the plots and the value obtained for HCl is more and close to unity (0.924). The values of  $K$  obtained from Flory–Huggins adsorption isotherm were substituted into Equation 3.11 to obtain values of free energy of adsorption which are recorded in Table 6. These values are negative and lower than the threshold value of  $-40\text{KJ/mol}$  required for chemical adsorption, hence the adsorption of the leaves extract of *Jatropha curcas* on mild steel is spontaneous, stable and favour the mechanism of physical adsorption. Also, the values of the size of parameters ( $x$ ) are positive as recorded in Table 6. This indicates that the adsorbed species of *Jatropha Curcas* leaves extract is bulky since it could displace more than one molecule of water from the mild steel surface.

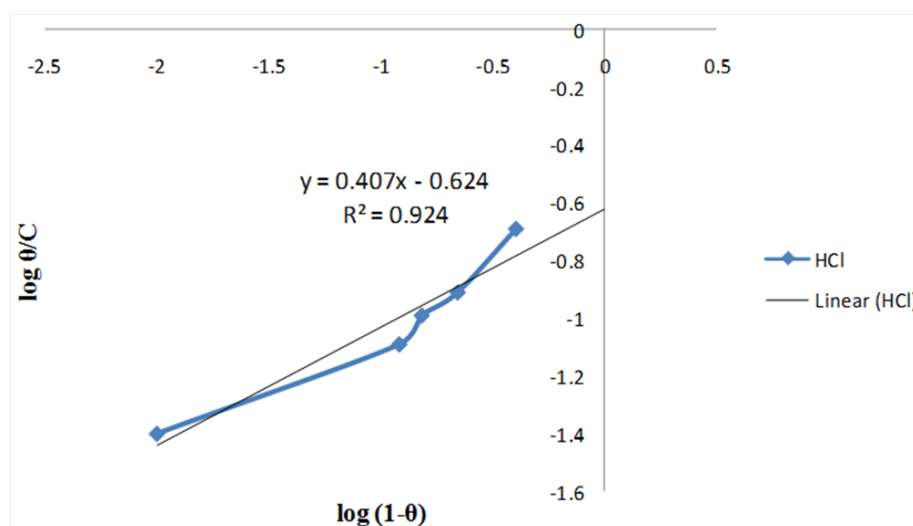


Figure 8. Flory–Huggins isotherm adsorption parameters for the adsorption of acid extract of leaves of *Jatropha Curcas* on the surface of mild steel.

Table 6 Flory–Huggins isotherm for adsorption of acid extract of *Jatropha* leaves extract on the mild steel

Adsorption Isotherm	$R^2$	Log $K$	$\Delta G_{\text{ads}}$ KJ/mol	$x$
Flory–Huggins	0.924	0.624	-13.78	0.407



Figure 9 and Table 7 show results obtained for the adsorption parameters when Freundlich isotherm model was used. The plot shows that the fitting of the isotherm does not give a perfect straight line which implies that the adsorption model of the leaves extract of *Jatropha* does not perfectly obey Freundlich adsorption isotherm as compared to the Langmuir parameters that gave a good fit of 0.999.

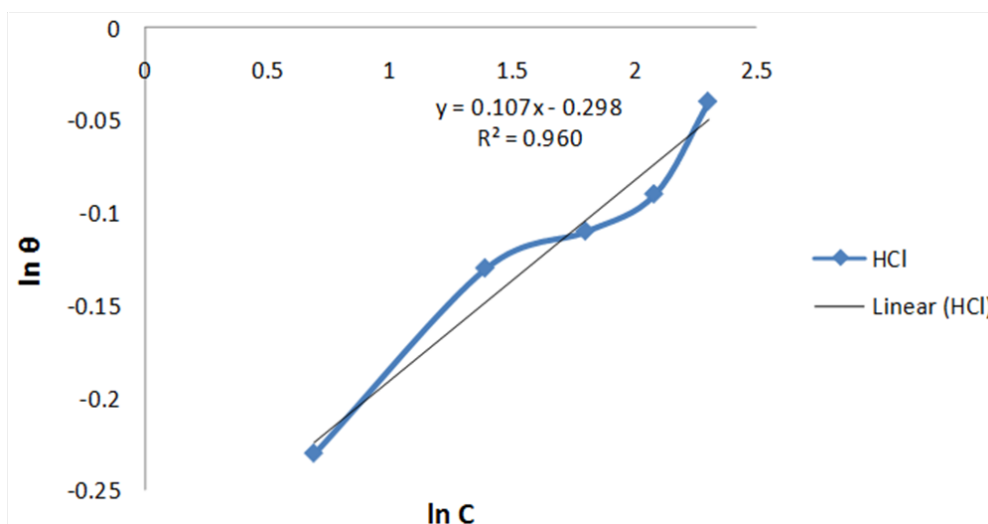


Figure 9. Freundlich isotherm for adsorption of *Jatropha Curcas* extract in HCl on mild steel surface

Table 7 Freundlich adsorption parameters for *Jatropha curcas* leaves extract in HCl solution

Adsorption Isotherm	$R^2$	K	$\Delta G_{ads}$ KJ/mol	n
Freundlich	0.960	0.742	- 9.40	0.107

#### 4. Conclusions

On the basis of this study, the following conclusions are drawn:

- I. The leaves extract of *Jatropha Curcas* acts as a good and efficient inhibitor for corrosion of mild steel in HCl medium.
- II. The inhibition of the corrosion of mild steel by acid extract of JC is due to the phytochemical constituents in the plant extract.

- III. The negative sign of free energy of adsorption indicates that the adsorption of *Jatropha curcas* leaves extract on mild steel surface was a spontaneous process and the mechanism of inhibition was found to be physisorption.
- IV. The adsorption of different concentrations of the *Jatropha curcas* leaves extract on the surface of mild steel in HCl solution fits into Langmuir adsorption isotherm model. However, Flory–Huggins and Freundlich isotherms were found not to be suitable.
- V. Results obtained in weight loss method were in good agreement with thermometric method.

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