



# **Inhibition of Mild Steel Corrosion by Leaves Extracts of *Commelina benghalensis* L In 1M HCl Solution**

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## **Authors' contributions**

*This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.*

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## **ABSTRACT**

The inhibition of mild steel corrosion in 1M HCl by leaves extracts of *Commelina benghalensis* L was investigated using weight loss and potentiodynamic polarization techniques at room (30 °C) and elevated temperatures of 40, 50, 60 and 70 °C. The results of weight loss technique showed that the inhibition efficiency increased with increase in extract concentration and exposure time but decreased with rise in temperature. The maximum inhibition efficiency obtained was 81.71 % at 0.5 g/L concentration of leaves extracts. Corrosion rates decreased with increased concentration of extracts but increased with increase in temperature. Potentiodynamic polarization method results indicated that *Commelina benghalensis* L is a mixed - type inhibitor as it modified both the anodic and cathodic reactions in 1M HCl solution. The adsorption of the extracts on the mild steel coupons

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was observed to follow Langmuir and Temkin adsorption isotherms. The negative values of Gibb's free energy of adsorption ranged from -21.35 to -23.03 kJ/mol, this suggests that inhibitor molecules were physically and spontaneously adsorbed onto the metal surface.

**Keywords:** Adsorption isotherm; *Commelina benghalensis* L; corrosion inhibition; inhibitor concentration; leaves extract; mild steel; weight loss.

## 1. INTRODUCTION

"Investigation of equipment and structural failures in the industries points to corrosion as the primary cause. Thus the failures of metallic mild steel structures is a direct consequence of corrosion which is a destructive attack on a metal by chemical or electrochemical reaction with its environment" [1]. Hence, the need to protect metallic structures from corrosion to enhance their performance cannot be overemphasized as corrosion can cause direct rusting of metal, resulting in loss of strength, ductility or cracking [2]. "In trying to combat corrosion, many methods have been used in recent times including the use of inhibitors which have been reported to be very effective in different environments" [3–5].

"A corrosion inhibitor is a chemical substance that when added to an environment in small quantities would lower the corrosion rate of a metal. As a result of research by scientists to find effective methods of controlling corrosion, there has been increasing interest in the area of green corrosion inhibitors. This has become necessary due to increasing environmental regulations to stop the use of toxic chemicals which are hazardous to the environment and mankind. The use of commercial inhibitors such as chromates and nitrates which can effectively control corrosion but are very expensive and poisonous to humans is now been substituted by green inhibitors which are non-toxic, cheap and cause no harm to the environment" [6].

Green corrosion inhibitors are simply made from extracts of plant materials, they are natural, cheap raw materials which are biodegradable, ecofriendly and do not contain heavy metals or other toxic substances. The use of plant materials as good corrosion inhibitors have been reported by many researchers [6,7–12].

"The inhibitive effect of plant materials is attributed to the presence of chemical compounds that are contained in them such as alkaloids, tannins, flavonoids, saponins, etc. These natural organic compounds are either synthesized or extracted from herbs, spices or

medicinal plants which have made plants a rich source of chemical constituents that can be used for corrosion inhibition" [13].

"The inhibitive effect of Cocoa leaves (*Theobroma cacao*) and Cola nut (*Cola acuminata*) extracts on the corrosion of mild steel in sea water at room temperature was investigated" by Umoru *et al.* [14]. The results obtained indicated that they are potential inhibitors of mild steel corrosion. Adzor *et al.* [15], investigated "corrosion inhibitive effect and adsorption behavior of hibiscus *sabdariffa* calyx on low carbon steel in acidic media and observed that it inhibited corrosion to 95 % efficiency". Nnanna *et al.* [16], carried out investigation on *Gmelina arborea* bark extract as a corrosion inhibitor for mild steel in acidic solution. The results showed that it acted as a good inhibitor with 94% efficiency. Iroha and Chidiebere [12] evaluated "the inhibitive effect of *Annona muricata* leaves extract on low-carbon steel corrosion in acidic media. The result was that the Leaves extract of *Annona muricata* functioned as effective corrosion inhibitor for low-carbon steel and the inhibition efficiency was concentration dependent as it increases with increase in concentration".

In the present study, the use of *Commelina benghalensis* L leaves extracts as corrosion inhibitors for mild steel in 1M HCl solution have been investigated using potentiodynamic polarization and gravimetric methods at room and elevated temperatures. *Commelina benghalensis* L (CBL) is commonly known as benghal dayflower or tropical spider wort. It belongs to a family of *Commelinaceae* comprising of over 500 species with distinct characteristics. The plant is native only to tropical Asia and Africa and it can be found in Nigeria, Ethiopia, Kenya, Senegal and Cameroon. It is a widely distributed herbaceous weed that usually invades agricultural sites and disturbed areas. This rapid growing plant is one of the most troublesome weeds for 25 crops in 29 countries; hence it is found on the International Union for Conservation of Nature (IUCN) red list of species [17]. It is used in treating diseases such as

swelling of skin, leprosy, anti-inflammatory effects, and as a laxative. The research will help find a beneficial application of an otherwise very troublesome weed.

## 2. MATERIALS AND METHODS

The materials used in this study are mild steel sheet of 0.5 mm thickness obtained from Makurdi main market, and *Commelina benghalensis* L leaves (CBL) which were collected from Antsah village, Northbank, Makurdi, Benue State, Nigeria. The materials were identified at the Forestry Department of Joseph Sarwuan Tarka University Makurdi (JOSTUM). Other materials used include distilled water, analytical grade HCl, acetone, filter paper, copper wire, digital weighing balance, beakers, measuring cylinders, desiccator, thermostatic digital water bath Searchtech HH-W420, mortar/pestle and Potentiodynamic Meter VERSASTAT 4 was used for potentiodynamic polarization experiment.

The mild steel sheet of composition (wt %): C (0.04), Si (0.004), Mn (0.225), P (0.004), Cu (0.002), Ti (0.007) and the rest Fe was mechanically cut into coupons of 40 × 30 × 0.5 mm. A small hole of 3 mm was drilled at the end of each coupon for its suspension in HCl solution with the help of flexible copper wire. The coupons were degreased, polished using different grades of silicon carbide impregnated with the emery paper of 120,400, 800 and 1000 grits; cleaned with acetone, dried and stored in a desiccator.

The leaves (Fig.1) were washed under running tap water and then separated into leaves parts. The separated materials were then dried under shade for four weeks to a constant weight and pounded in a mortar to reduce it to smaller sizes

which were later blended to powder form and stored for subsequent use.

The extracts were prepared from dried leaves powdered materials at the biological laboratory Joseph Sarwuan Tarka University, Makurdi (JOSTUM) using cold extraction process as stated by Umoren et al. [18]. Fifty grams (50 g) of the prepared material was weighed using a digital weighing scale and soaked in 500mls of absolute ethanol for 48 hours with occasional agitation. The solution was double filtered with cheese cloth and the filtrate collected in a conical flask. The extract was dried using water bath at 45 °C and collected in a sample bottle and stored in a refrigerator at 4 °C for subsequent use.

The acid solution was prepared by diluting 83.61 ml of concentrated Hydrochloric acid of 37 % purity in distilled water to make 1M HCl solution. To achieve this, 83.61 ml of HCl was diluted with 900 ml of distilled water and made up to 1000 ml of HCl solution. From this volume of 1000 ml, 200 ml was measured and used as the corrosive media in six 250 ml beakers. Inhibitor concentrations of 0.1, 0.2, 0.3, 0.4 and 0.5 g/L were then prepared. 0.5 g of each extract was added to 200 ml of HCl solution to give the stock solution with concentration of 2.5 g/L.

The different working concentrations of (0.1 to 0.5 g/L) were obtained using the dilution principle as shown in equation 1.

$$C_1V_1 = C_2V_2 \quad (1)$$

where  $C_1$  is the initial concentration of the extract,  $V_1 = \frac{C_2V_2}{C_1}$  is the initial volume needed to obtain desired concentration,  $C_2$  is the final concentration (working concentration) and  $V_2$  is the final volume (working volume).



Fig. 1. *Commelina benghalensis* L leaves

The polished coupons were then weighed using a digital weighing balance. These weighed coupons were then fully immersed in six beakers of 250 cm<sup>3</sup> containing 0.1 to 0.5 g/L extract concentration in 200 cm<sup>3</sup> HCl solution. The sixth beaker contained only the acid solution without inhibitor in it. Two coupons were immersed in each beaker containing the media. Weight loss measurements were carried out after every 24hrs of immersion progressively for 168 hours at room temperature. The coupons were removed from the solution after 24 hours; washed with distilled water, cleaned with acetone, dried in air and reweighed and average weight loss recorded. This procedure was repeated to test the coupons at elevated temperatures of 40, 50, 60 and 70 °C but with replacement of coupons and exposure time of 3 hours in a thermostatic water bath capable of temperature regulation. The weight loss measurements were obtained according to ASTM G31-72, which is the standard methodology for this technique in the laboratory.

The corrosion rates of mild steel in 1.0 M HCl solutions in the absence and presence of *Commelina benghalensis* plant extract were determined at room and elevated temperatures using weight loss as expressed by equation (3). The difference in weight of the coupons before and after exposure in acidic solution is recorded as weight loss (W) of the metal as presented in equation 2 [9].

$$W = W_o - W_i \quad (2)$$

where, W = weight loss in grammes (g), W<sub>o</sub> = weight before immersion (g) and W<sub>i</sub> = weight after immersion (g).

The corrosion rate (C.R), inhibition efficiency (I.E) and degree of surface coverage were then calculated using equations 3, 4 and 5 respectively.

$$C.R. = \frac{87.6W}{DAT} \quad (mm/yr) \quad (3)$$

where w = weight loss in (g), D = density of the metal, (g/cm<sup>3</sup>), A = total surface area of the coupon (cm<sup>2</sup>), 87.6 is the unit conversion constant, and T = time of exposure (hours).

$$I.E. = \frac{CR_o - CR_1}{CR_o} \times 100 \% \quad (4)$$

where I.E. = inhibition efficiency (%), CR<sub>o</sub> and CR<sub>1</sub> are corrosion rates in the absence and presence of inhibitor respectively.

$$\theta = \frac{CR_o - CR_1}{CR_o} \quad (5)$$

where  $\theta$  is the degree of surface coverage, CR<sub>o</sub> and CR<sub>1</sub> are corrosion rates in the absence and presence of inhibitor respectively.

The influence of temperature on the corrosion behavior of mild steel in 1M HCl in the absence and presence of the various inhibitor extracts was also investigated by weight loss method at 40, 50, 60 and 70 °C. Twelve (12) pre-cleaned mild steel coupons were weighed separately on analytical digital weighing balance and each suspended with a string and two immersed in each beaker. 0.1 – 0.5 g/L, extract concentrations, while the sixth beaker contained 1M HCl solution without inhibitor as a control. The six beakers with and without inhibitor solutions containing the coupons were kept at the desired temperature in a thermostatic water bath for a period of three (3) hours. At the end of this time, the specimens were carefully removed, washed in doubly distilled water, dried and reweighed. The weight loss was recorded for the purpose of calculating the corrosion rate due to effect of elevated temperature using equation (3).

Potentiodynamic polarization investigations were conducted in the absence and presence of the plants extracts to evaluate its effect on the corrosion potential ( $E_{corr.}$ ), corrosion current density ( $I_{corr.}$ ) and the anodic ( $\beta_a$ ) and cathodic ( $\beta_c$ ) Tafel constants.

The electrochemical experiments were performed using a computer controlled potentiostat instrument, VERSASTAT 4. The set up consisted of a three – electrode corrosion cell made up of mild steel coupons as the working electrode (WE), with surface area of 1cm<sup>2</sup>; saturated silver/silver chloride as reference electrode, and platinum rod as counter electrode. The test electrolyte was 1M HCl solution in the presence and absence of CBL extracts in varying concentrations of 0.1, 0.2, 0.3, 0.4 and 0.5 g/L and the blank.

All electrochemical experiments were conducted at room temperature (30 ± 2 °C) using 100 ml of the inhibitor solution in a stationary condition. The working electrode was immersed in a test solution for 30 minutes until a stable open-circuit potential (OCP) was attained for corrosion to reach a steady /equilibrium state.

Polarization measurements were carried out at a scan rate of 1.0 mV/s at a potential initiated at -250 mV to +250 mV. The test sample and electrolyte were replaced after each experiment. The linear portion of the Tafel plot of the anodic and cathodic curves were extrapolated to obtain the corrosion current densities ( $I_{corr}$ ) and corrosion potential ( $E_{corr}$ ), Anodic Tafel slope ( $\beta_a$ ) and cathodic Tafel slope ( $\beta_c$ ) were determined from the experimental curve. The inhibition efficiency was calculated using the values of current densities in the absence and presence of inhibitor using equation (6).

$$IE_{corr} = \frac{I_{corr}^0 - I_{corr}}{I_{corr}^0} \times 100 \text{ (%)}$$
 (6)

Where,  $I_{corr}^0$  and  $I_{corr}$  are the current density values in the absence and presence of inhibitor respectively.

### 3. RESULTS AND DISCUSSION

The results arising from the gravimetric analysis obtained from variation of weight loss with exposure time for mild steel coupons immersed in the absence and presence of CBL extracts were used in calculation of corrosion rates. In Fig. 2, it can be observed that corrosion rate decreased remarkably with addition of various inhibitor concentrations compared to the blank solution for CBL inhibitors. The reduction in corrosion rate can be seen to be reflected in the inhibition efficiency, which increases with increase in extract concentrations as presented in Fig. 2 reaching the maximum of 78.2 % for CBL extracts.

Fig. 3 show that there is increase in inhibition efficiency as the concentration increases. This increase in inhibition efficiency with concentration could be attributed to adsorption of more organic materials on the metal surface which creates a barrier for mass and charge transfer thereby reducing corrosion process. Increasing inhibitor concentration increases surface coverage and reduces available sites for corrosion reaction, thus reducing corrosion rate [5,16].

The effect of temperature on corrosion inhibition of mild steel in the absence and presence of (0.1 to 0.5 g/L) CBL extracts were investigated for 3 hours at temperature range of 40 °C -70 °C. Fig. 4 show the effect of temperature on corrosion rate of mild steel in the absence and presence of CBL extract. The corrosion rate increases with temperature but decreases as the inhibitor concentration increases. The increase in corrosion rate is due to the fact that corrosion process is enhanced at higher temperatures as a result of increased movement of inhibitor molecules which weakens their adsorption capability [2]. This affects the inhibition efficiency which decreases with increase in temperature at various extract concentrations as presented in Fig. 5. This finding is in agreement with the findings of Ebenso et al. [3], Solomon et al. [19] and Ekanem et al. [20]. The dependence of corrosion rate on temperature can be expressed using Arrhenius equation 7.

$$\text{Log} \frac{\rho_2}{\rho_1} = \frac{E_a}{2.303R} \left( \frac{1}{T_1} - \frac{1}{T_2} \right)$$
 (7)

where  $\rho_1$  and  $\rho_2$  are the corrosion rates at temperatures  $T_1$  and  $T_2$  respectively,  $E_a$  is the activation energy and  $R$  is the gas constant

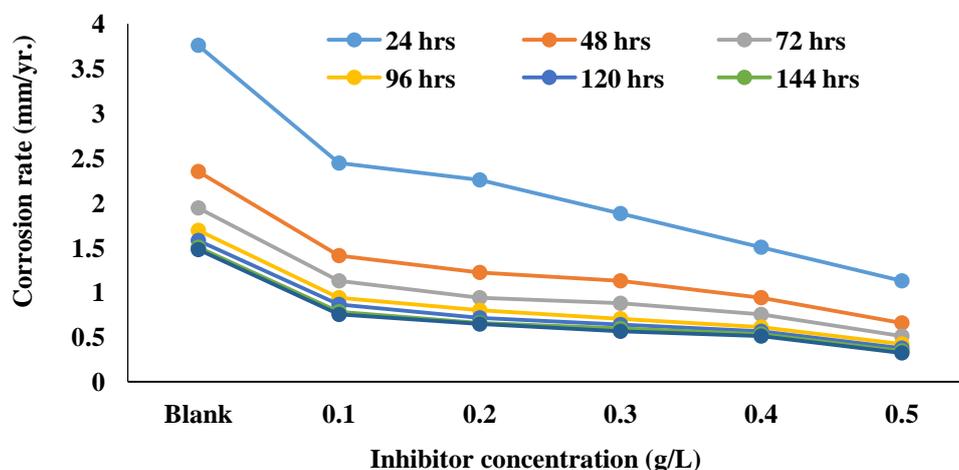


Fig. 2. Corrosion rate against CBL inhibitor concentration in 1M HCl solution

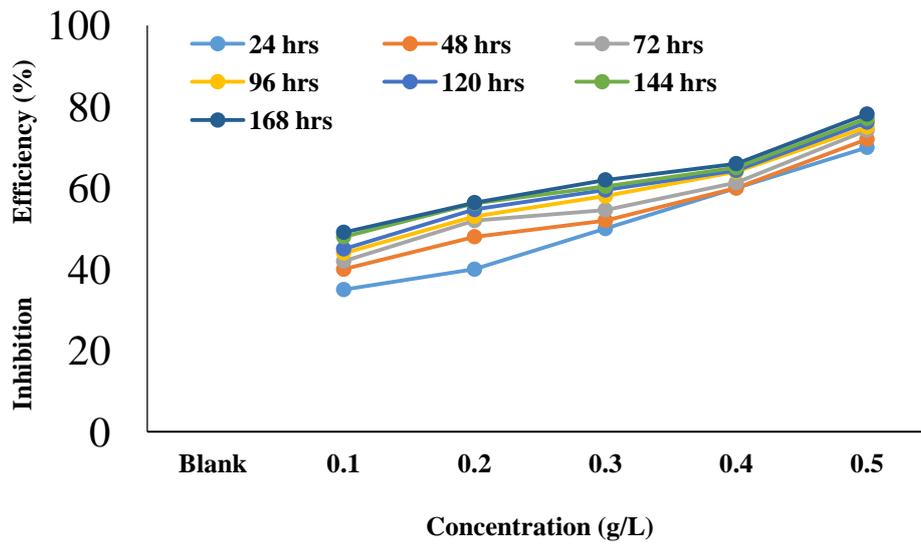


Fig. 3. Inhibition efficiency against CBL inhibitor concentration at room temperature (30 ± 2)

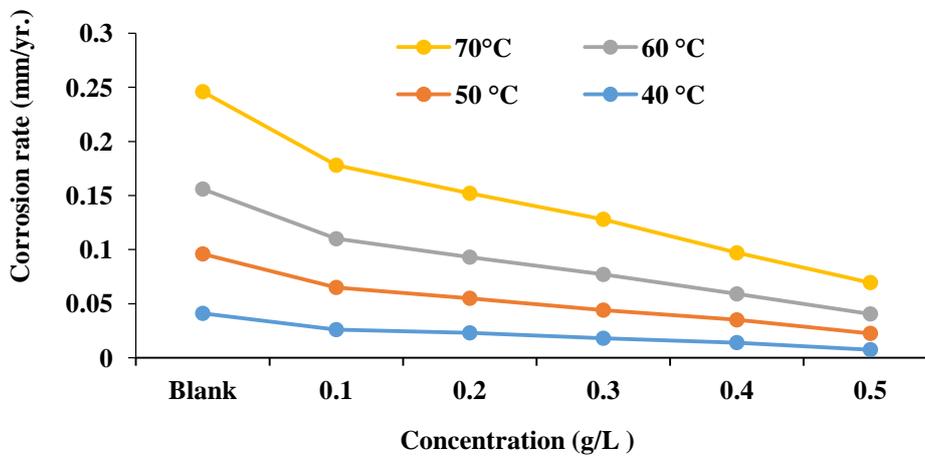


Fig. 4. Variation of corrosion rate with different CBL extract concentrations and temperatures

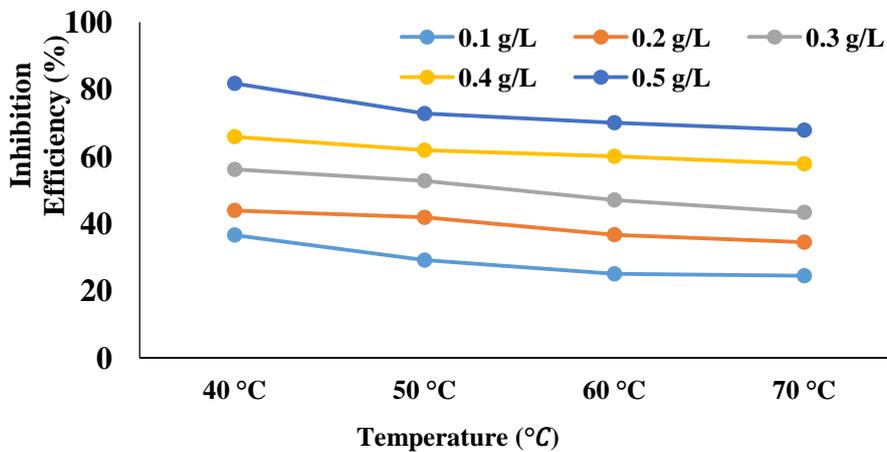


Fig. 5. Variation of inhibition efficiency of CBL extract with different concentrations and temperatures after 3 hours of immersion

The results of potentiodynamic polarization experiments are presented in Table 1 and Fig. 6. The electrochemical parameters such as the corrosion potential ( $E_{corr}$ ), corrosion current density ( $I_{corr}$ ) and the anodic ( $\beta_a$ ) and cathodic ( $\beta_c$ ) Tafel constants were collected from Tafel plots. It can be observed from the polarization parameters that in the presence of the inhibitors, there is a decrease in anodic and cathodic current densities and the corrosion potential ( $E_{corr}$ ) for the extract concentrations compared to the uninhibited. This implies that the extracts have inhibitory effects on the mild steel corrosion [21]. The higher values of ( $\beta_c$ ) compared to those of ( $\beta_a$ ) shows that the cathodic reaction was the predominant factor and that inhibitors acted as mixed type but with more cathodic influence [12].

The consideration of adsorption isotherm gives us the idea of how the inhibitor molecules are adsorbed on the metal surface through their polar functions. Adsorption isotherm values are used to explain the mechanism of corrosion inhibition of organo-electrochemical reactions.

In this study, Langmuir and Temkin isotherms were investigated as presented in Figs. 6 and 7. Langmuir isotherm is given by the expression (8).

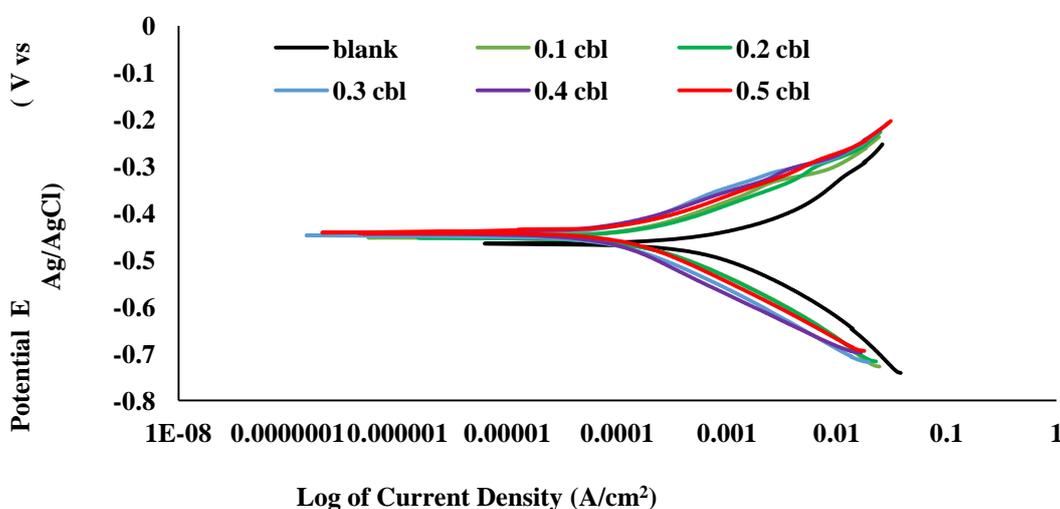
$$\frac{C}{\theta} = \frac{1}{K_{ads}} + C \tag{8}$$

where  $C$  = inhibitor concentration,  $\theta$  = degree of surface coverage, and  $K_{ads}$  is the adsorptive equilibrium constant.

The plots of  $C/\theta$  against  $C$  are given in Fig. 7 for CBL. Linear plots were obtained with good correlation coefficient of ( $R^2$ ) close to unity which suggests that the adsorption of this inhibitor obeys Langmuir isotherm model. The adsorptive equilibrium constant  $K_{ads}$  decreases with increase in temperature of the environment (Table 2) which indicates that the interactions between the adsorbed molecules and the metal surface become weaker at higher temperatures and could lead to desorption of inhibitor from the metal surface [22].

**Table 1. Potentiodynamic polarization parameters for mild steel in the absence and presence of various concentrations of *Commelina benghalensis* Leaf (CBL) extract as inhibitor**

Inhibitor concentration	$E_{corr}$ (mV)	$I_{corr}$ ( $\mu$ A)	$\beta_a$ (mV)	$\beta_c$ (mV)	Corrosion rate (mm/yr.)
Blank	-464.679	-938.44	126.634	158.208	10.889
0.1 g/L	-451.107	-181.076	97.029	118.51	2.1011
0.2 g/L	-450.741	-156.843	82.438	107.076	1.8200
0.3 g/L	-446.095	-86.96	92.485	110.477	1.0091
0.4 g/L	-443.49	-74.278	71.121	108.424	0.8619
0.5 g/L	-440.613	-51.337	46.496	63.83	0.5957



**Fig. 6. Potentiodynamic polarisation curves for mild steel corrosion using various concentrations of CBL extract**

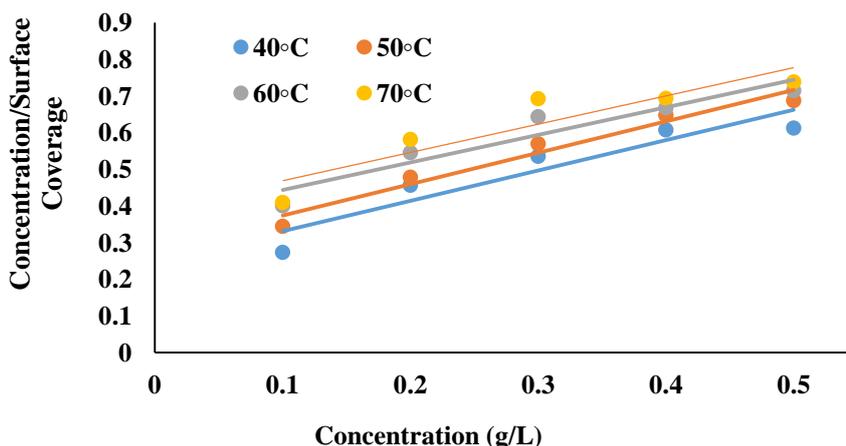


Fig. 7. Langmuir adsorption isotherm for mild steel in 1 M HCl solution in the presence of CBL extract

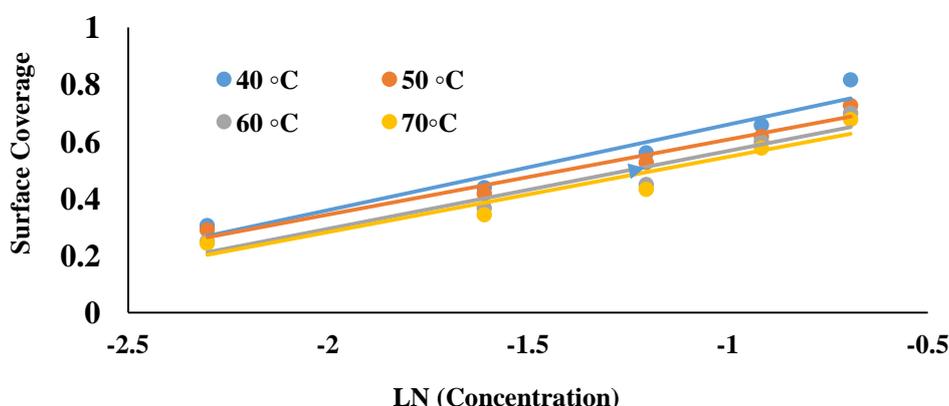


Fig. 8. Temkin adsorption isotherm for mild steel in 1.0 M HCl solution in the presence of *Commelina benghalensis* leaf (CBL) extract

The plots of surface coverage against  $\ln(C)$  obtained from the expression (9)

$$\theta = \left(\frac{1}{f}\right) K_{ads} C \quad (9)$$

where  $f$  = factor that determines adsorbent - adsorbates interaction,  $\theta$  = the degree of surface coverage,  $K_{ads}$  is the equilibrium constant and  $c$  = concentration of inhibitor [23]; gives straight lines as shown in Fig. 7 indicating that Temkin isotherm is also obeyed by CBL extracts with  $R^2$  values close to unity.

The calculated values of activation energy ( $E_a$ ) and heat of adsorption ( $Q_{ads}$ ) are presented in Table 2 and it can be observed that  $E_a$  values are higher in the presence of inhibitor than in its absence. This may be attributed to the geometric

blocking effect of adsorbed species on the metal surface and it is suggestive of physical adsorption, while lower values of  $E_a$  suggests chemisorption [19,24].

The heat of adsorption ( $Q_{ads}$ ) values were calculated from the variation of surface coverage with temperature as expressed in equation (10);

$$Q_{ads} = 2.303R \left[ \log \left( \frac{\theta_2}{1-\theta_2} \right) - \left( \frac{\theta_1}{1-\theta_1} \right) \right] \quad (10)$$

Where,  $\theta_1$  and  $\theta_2$  are the degrees of surface coverage at temperature  $T_1$  and  $T_2$  respectively.

Table 3 shows negative values of  $Q_{ads}$  which implies spontaneous adsorption of the inhibitor and it is indicative of physisorption, while positive values are supportive of chemisorption mechanism of adsorption [18].

**Table 2. Adsorption parameters for CBL inhibitors at various temperatures from langmuir and temkin isotherm models**

Temp. (°C)	K <sub>ads</sub> (L/mol.)	ΔG <sup>0</sup> <sub>ads</sub> (kJ/mol)	R <sup>2</sup> (Langmuir)	R <sup>2</sup> (Temkin)
40	5,770	- 22.54	0.8741	0.9406
50	4,098	-22.27	0.9614	0.9663
60	3,333	-22.46	0.9048	0.9311
70	3,210	-23.03	0.8416	0.9212

**Table 3. Calculated values of activation energy and heat of adsorption at 40°C and 70°C for *Acanthospermum hispidum* leaf (CBL) extract concentrations**

Extract Conc. (g/L)	E <sub>a</sub> (kJ/mol)	Q <sub>ads</sub> (kJ/mol)
Blank	23.40	-
0.1	28.58	-17.20
0.2	28.03	-12.00
0.3	30.97	-15.26
0.4	29.72	-10.14
0.5	40.25	-22.36

#### 4. CONCLUSION

The leaves extracts of *Commelina benghalensis* L investigated were found to be effective in inhibition of mild steel corrosion in 1M HCl solutions. The inhibition was achieved through physical adsorption of the extracts on the mild steel surface according to Langmuir and Temkin isotherm models. The corrosion rate was found to decrease with increase in inhibitor concentration at room temperature but increased at elevated temperatures, while inhibition efficiency increased with increase in concentration but decreased with rise in temperature. The thermodynamic parameters of activation energy (E<sub>a</sub>), heat of adsorption (Q<sub>ads</sub>) and Gibbs free energy of adsorption (ΔG<sub>ads</sub>), suggests that there was spontaneous adsorption of inhibitors on the metal surface and the negative values of Q<sub>ads</sub> and ΔG<sub>ads</sub> are supportive of physical adsorption mechanism. Potentiodynamic polarization parameters obtained show that inhibitors of CBL acted as mixed –type by modifying both the anodic and cathodic reactions.

#### COMPETING INTERESTS

Authors have declared that no competing interests exist.

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