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Research Article

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Inhibition potentials of *Solanum macrocarpon* leave extract as a green corrosion inhibitor of mild steel in an acidic solution

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Abstract

The adsorption and inhibitive properties of methanol extract of leaves of *Solanum macrocarpon* as green inhibitor for the corrosion of mild steel in 2M H₂SO₄ acid solutions were studied using gasometric techniques at 303K, 313K, and 323K, respectively. The inhibition efficiency increased with concentration and declined when the temperature rose. The results indicated that the methanol extract of leaves of *Solanum macrocarpon* is a suitable corrosion inhibitor for the inhibition process as 91% maximum inhibition efficiency was obtained at 0.5%w/v extract concentration at 303K. The plant extract's inhibition potential was ascertained due to the presence of organic compounds which contain heteroatoms in their functional groups. Thermodynamics parameters (E_a , Q_{ads} , and ΔG_{ads}) demonstrated that the corrosion mechanism was by physical adsorption. The adsorption process was found to obey Langmuir, Temkin, Freundlich, and El-Awardy ($R^2 > 0.9$). The results showed that *Solanum macrocarpon* methanol extract could be a cost-friendly corrosion inhibitor in industries.

Keywords: *Solanum macrocarpon*, corrosion, inhibition, Mild steel, sulphuric acid, adsorption.

Introduction

Corrosion is a chemical process that involves the deterioration of materials with their environment [1,2]. It is a gradual process that destroys the materials leading to economic waste. For industrial-related tasks, mild steel and hydrochloric acids are most frequently utilized in industries. As a result, scientists are now researching how corrosion inhibitors affect mild steel in an acidic environment [3,4,5]. The cleaning and descaling of metals in the industries is done with acid solutions, which require corrosion inhibitors to prevent corrosion of the metals [6,7]. Inhibitors are chemical substances that help mitigate the corrosion rate of materials [8,9]. Inorganic inhibitors have been used for decades to mitigate the spread of materials' corrosion in industries [6]. Inorganic inhibitors have declined due to their toxicity, high cost, and non-ecofriendly [9,10].

Recently, plant extract has become interesting research in corrosion control due to its eco-friendly nature, non-toxic, and affordability. Several works have used plant extract as eco-friendly inhibitors [3,4,11,12]. This plant extract contains phytochemical compounds with heteroatoms in their functional groups [13,14,15]. These heteroatoms (N, S, O, and P) are active centers of the adsorption process [13]. The mechanism of the reaction is that the heteroatoms have lone pair electrons, which facilitate the transfer of electrons between the inhibitor and the metal surface, thereby forming a compact barrier and reducing corrosive attack [14,15].

This work advances our efforts to support the current interest in environmentally friendly corrosion inhibitors; the present study investigated and evaluated the inhibitive property of methanol extract of *Solanum macrocarpon* (garden egg) leaves as a green inhibitor for mild steel corrosion using the gasometric method.

2.0 Material and Methods

2.1 Materials

Each coupon was physically pressed and cut from a sheet of mild steel that measured 4.0 by 3.0 by 0.12 cm. A tiny hole of diameter 3mm was drilled on each coupon close to its upper edge to suspend it in the corrosive medium. Each coupon underwent ethanol cleaning, acetone rinsing, and air drying procedures before being preserved in a desiccator [16]. Analytical grade reagents and chemicals purchased from Sigma –Aldrich, USA, were used for all the studies and were prepared using distilled water. 2M concentration of H_2SO_4 was prepared and utilized for the study.

2.2 Extraction of plant leaves and preparation of extracts

Leaves of *Solanum macrocarpon* were sourced from a local market. The leaves were washed with clean water, shade-dried, ground into powder, and extracted using methanol by the soxhlet extraction technique. 10g was used to make various extract concentrations (0.1-0.5% w/v) in 2M H₂SO₄ solution for the experiment.

2.3 Gasometric experiment

A gasometric assembly was used for this experiment and followed the method [17]. The equipment consists of a volumetric flask attached to a burette through a flexible 10mm air pipe. The burette was filled to a specific volume using paraffin oil. 100 mL of 2M H₂SO₄ was added to the volumetric flask during the monitoring of corrosion experiments using this method, and the initial volume of air in the burette was recorded. Then, the flask was swiftly sealed with a glass stopper after a mild steel coupon with dimensions of 4.0 x 3.0 x 0.12cm and an existing weight was placed into the acid solution (blank). Every five minutes for 25 minutes, volume changes in the paraffin oil in the graded burette were used to measure the amount of hydrogen gas that resulted from the corrosion reaction. In a different experiment, new coupons were submerged in a flask of corrodent while each extract from *Solanum macrocarpon* leaves was added at a different concentration (0.1, 0.2, 0.3, 0.4, and 0.5%w/v, respectively). The volume of hydrogen gas that evolved from each sample was recorded at the same time intervals as the blank. With the aid of a thermostat water bath, the experiment was carried out at 303, 313, and 323K. Based on the volume of hydrogen evolution, parameters such as inhibition efficiency and corrosion rate were calculated as follows [12].

$$\text{Inhibition efficiency (IE \%)} = \frac{V_0 - V_i}{V_0} \times \frac{100}{1} \quad (1)$$

V_0 and V_i are volumes of hydrogen evolved for the uninhibited and inhibited solutions, respectively.

$$\text{Surface coverage } (\Theta) = \frac{V_0 - V_i}{V_0} \quad (2)$$

where V_0 and V_i are volumes of hydrogen evolved for the uninhibited and inhibited solutions, respectively.

The rate of hydrogen evolution was evaluated using the equation below;

$$\text{Corrosion rate } CR_H (\text{cm}^3/\text{min}) = \frac{V_t - V_i}{t_t - t_i} \quad (3)$$

V_t and V_i are volumes of hydrogen gas evolved at time intervals t_t and t_i , respectively.

2.4 Kinetic and thermodynamic parameters

Using the Arrhenius equation, the reaction's activation energy was determined [18].

$$\log_{10} \frac{CR_2}{CR_1} = \frac{E_a}{2.303R} \left(\frac{1}{T_1} - \frac{1}{T_2} \right) \quad (4)$$

where, correspondingly, T_1 (323K) and T_2 (303K) are the temperatures at which mild steel corrodes at the fastest rates (CR_1 and CR_2). The reaction's activation energy is designated as E_a is the activation energy, and R stands for the molar gas constant (8.314kJ/mol).

The following equation was used to compute the heat of adsorption [19].

$$Q_{ads} = 2.303R \left[\log \left(\frac{\theta_2}{1 - \theta_2} \right) - \log \left(\frac{\theta_1}{1 - \theta_1} \right) \right] \times \left[\frac{T_1 \times T_2}{T_2 - T_1} \right] \text{kJmol}^{-1} \quad (5)$$

Where Θ_1 and Θ_2 are the degrees of surface coverage of the inhibitor at the temperatures T_1 (303K), and T_2 (323K), respectively, and R stands for the molar gas constant.

3.0 Results and Discussions

3.1 Hydrogen evolution data

The volume of hydrogen evolved for the uninhibited and inhibited solutions at 30°C, 40°C, and 50°C is shown in Figures 1, 2, and 3. The amount of hydrogen gas released was seen to increase with longer immersion times but decrease with higher inhibitor concentrations. Moreover, the result showed that the mild steel corroded in 2M H₂SO₄ solution, but the presence of the inhibitor in the acid decreased the extent of metal degradation due to the shielding effect of the extract on the metal surface [15]. Similar results were observed by (Eddy *et al.* 2010). Hence, *Solanum macrocarpon* leaf extract can serve as an excellent green inhibitor for mild steel corrosion in 2M H₂SO₄ solution.

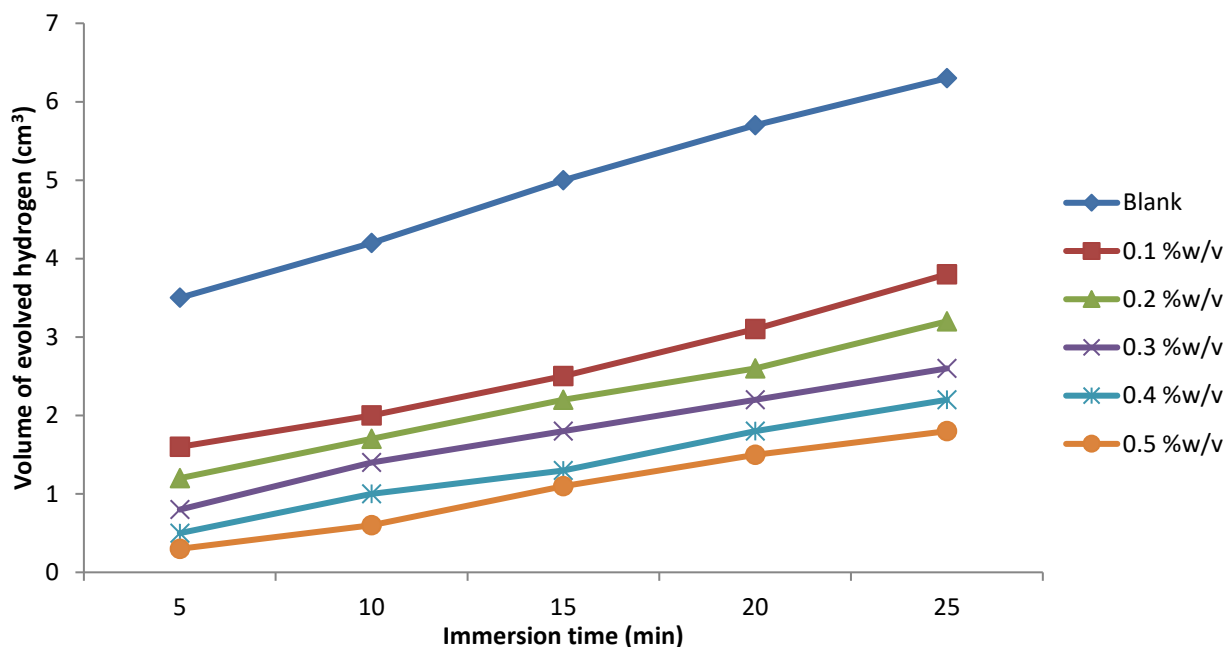


Fig 1: The volume of evolved hydrogen with immersion time for different inhibitor concentrations at 303K

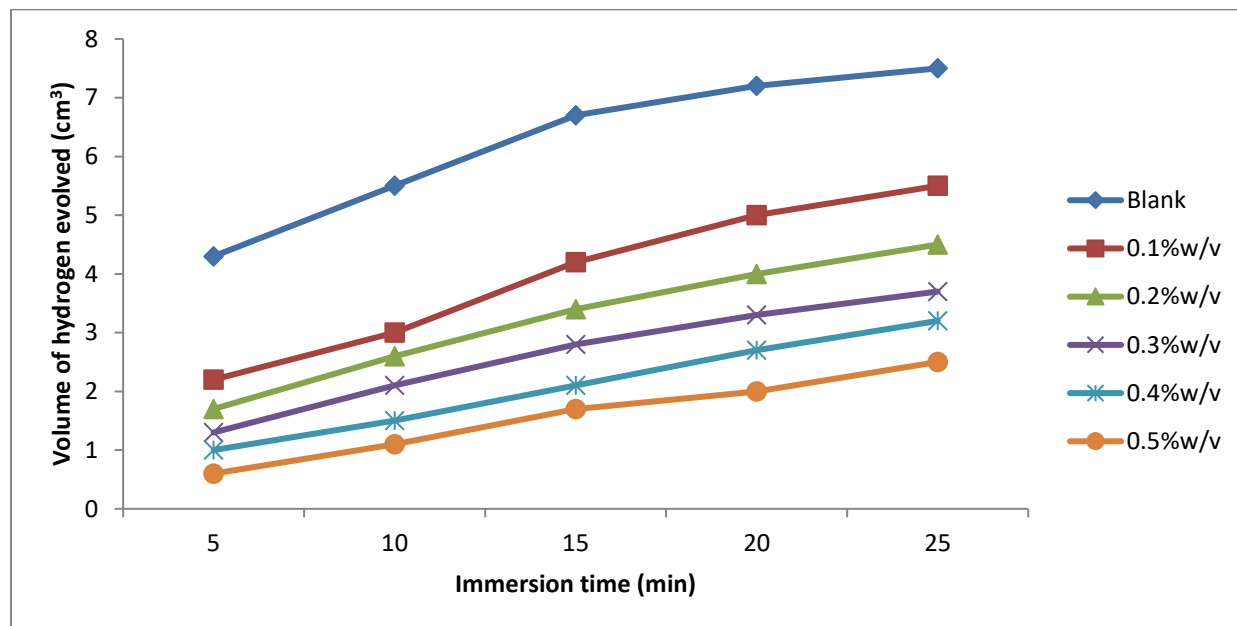


Fig. 2: The volume of evolved hydrogen with immersion time for different inhibitor concentrations at 313K

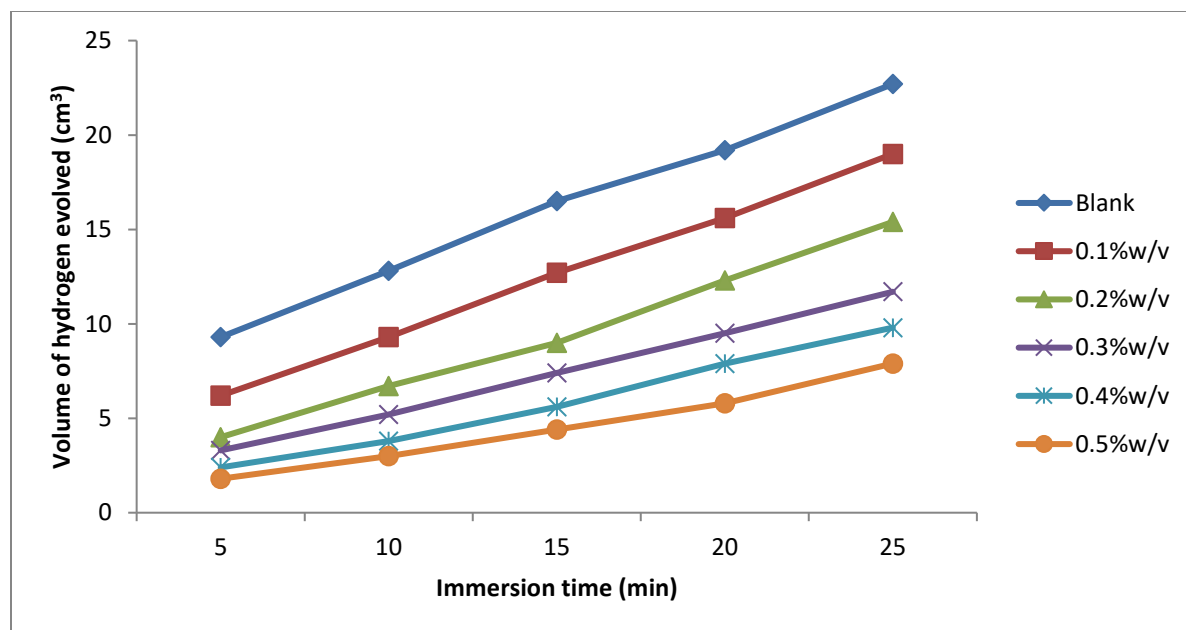


Fig. 3: The volume of evolved hydrogen with immersion time for different inhibitor concentrations at 323K

The effect of temperature on the volume of evolved hydrogen gas is illustrated using Fig 2.

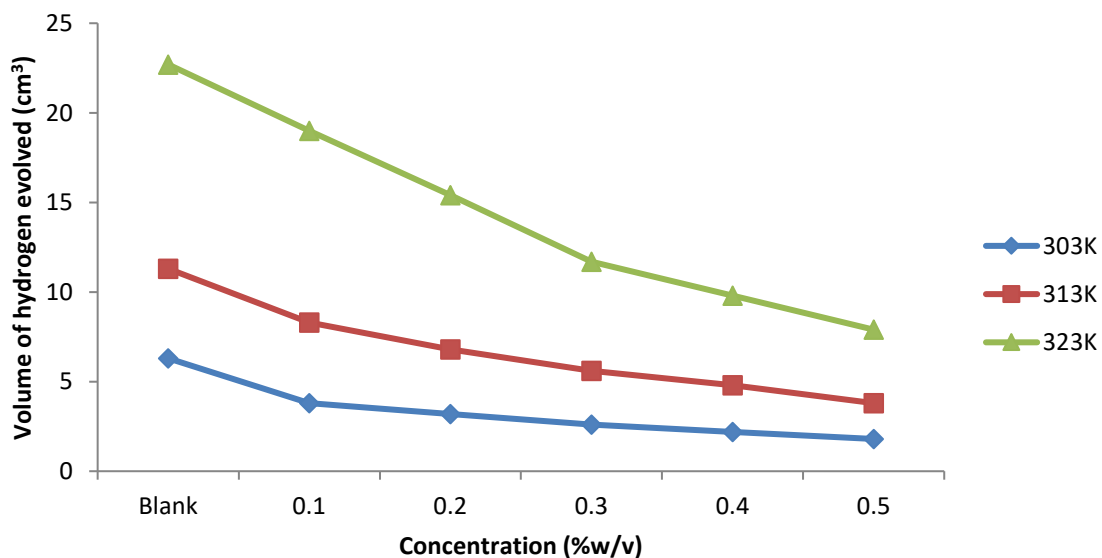


Fig. 4: The volume of evolved hydrogen with inhibitor concentrations at varying temperatures

The results also showed that the volume of evolved hydrogen gas increases with temperature rise due to the vulnerability of the metal to corrosion [2]. The inhibition potential of the inhibitor was greatly affected by temperature rise. Similar conclusions have been established [21].

3.2 Inhibition efficiency data

The variation of inhibition efficiency with the concentrations of the extract in 2M H₂SO₄ acid solutions at different temperatures was studied, as illustrated in Figure 5.

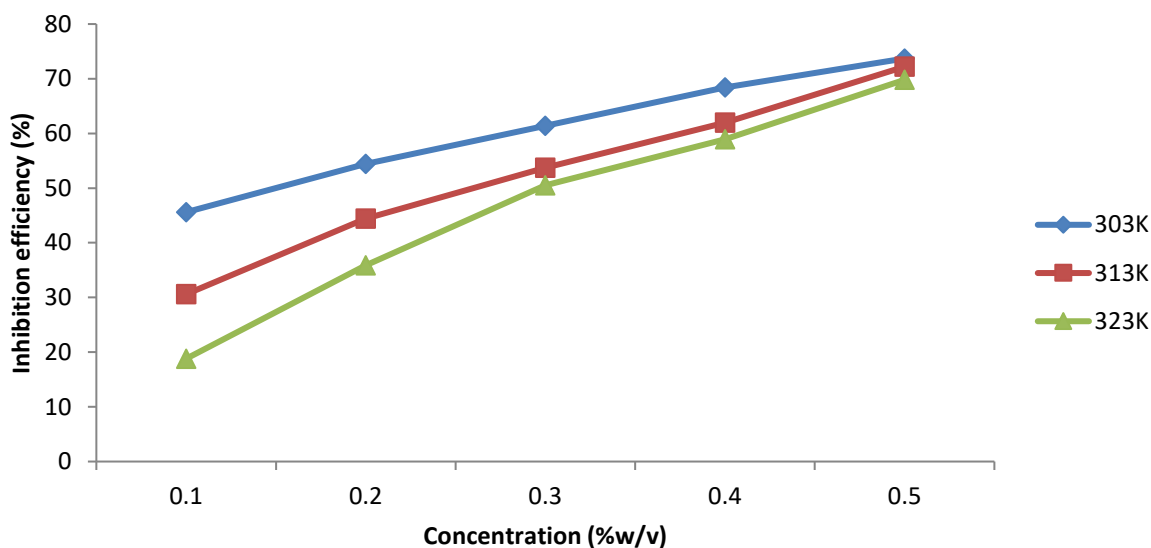


Fig. 5: Variation of inhibition efficiency with inhibitor concentration and temperature

The results showed that the extract's inhibition efficiency increases with the concentration of the extract in the acid solution. This suggested that as the extract concentration increased, so did the number of its molecules that were adsorbed onto the metal surface. This led to an increase in the percentage of the metal surface that these inhibitor molecules covered, preventing the acid's aggressive anions from continuing to corrode the metal. Other researchers have made similar conclusions [12]. As the temperature rises, the extract's ability to prevent corrosion declines due

to the increase in kinetic energy of the inhibitors, which reduces the adsorption strength at the binding sites between the inhibitor and the mild steel [17]. Hence the mode of inhibition was ascribed to physical adsorption. A similar result was observed by [17,20,22].

3.3 Corrosion rate data

Figure 6 shows the corrosion rate of the mild steel in different concentrations of the extract at different temperatures.

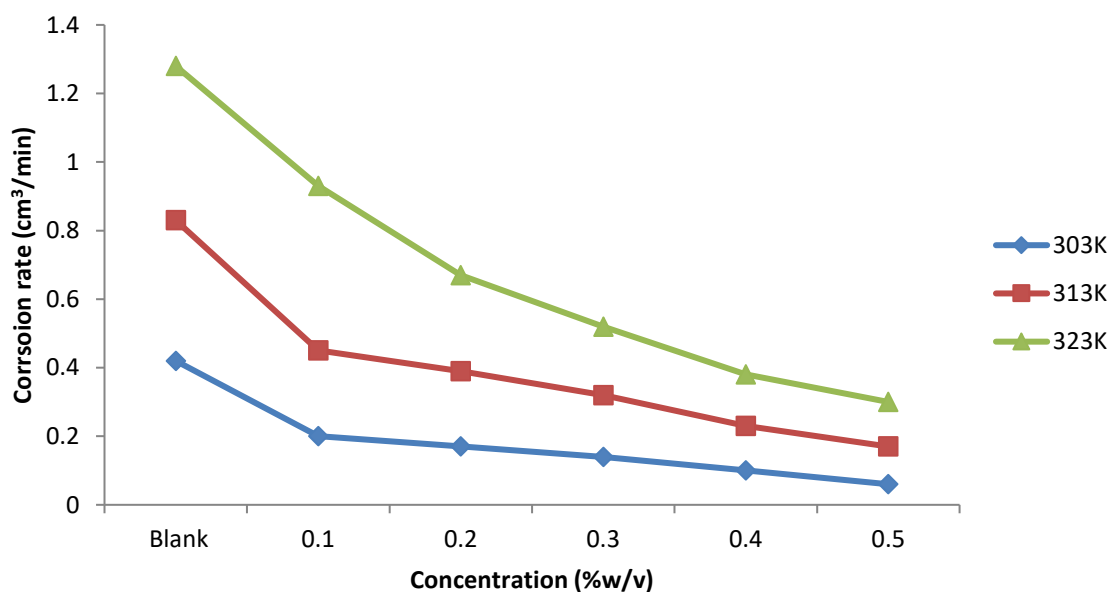


Fig.6: Variation of Corrosion rate with inhibitor concentration at varying temperatures

From the results obtained as depicted in the graph, the corrosion rate was found to be retarded by the presence of *Solanum macrocarpon* leaves extract as an inhibitor in the acidic medium, as it decreases progressively with an increase in extract concentration. Many researchers drew a similar conclusion [12,16,20]. As temperature rises, mild steel corrosion rates in both inhibited and uninhibited solutions both gradually increase, which may be due to faster reaction kinetics at both the anodic and cathodic sites [20].

3.4 Kinetic and thermodynamic data

The results of activation energy and heat of adsorption are shown in Table 1. The activation energies (E_a) varied from 57.64 - 62.99 kJmol⁻¹. The values are greater than the values of 46.36 kJmol⁻¹ obtained for the blank, showing that the extract slowed the rate of corrosion. The activation energies were < 80kJmol⁻¹ required for chemisorption [23]. Therefore the adsorption of the *Solanum macrocarpon* extract on a mild steel surface obeys the physical adsorption mechanism [23].

Table 1: The values of activation energy and heat of adsorption

Concentration (%w/v)	E_a (kJ/mol)	Q_{ads} (kJ/mol)
Blank	46.36	-
0.1	62.99	-47.51
0.2	58.99	-21.77
0.3	57.64	-16.74
0.4	62.02	-16.62
0.5	61.87	-15.73

Table 1 shows the heat of adsorption (Q_{ads}) values. The values were negative and ranged from -15.73 to -47.51 kJmol⁻¹, demonstrating the exothermic nature of the adsorption [12].

3.6 Adsorption Isotherm data

The obtained Θ values were applied to different adsorption isotherms such as Langmuir, Temkin, Freundlich, and El-Awardy. Equation 6 shows the Langmuir isotherm [19,23].

$$\frac{C}{\Theta} = \frac{1}{K} + C \quad (6)$$

C = inhibitor concentration, K = adsorption equilibrium constant, and Θ = surface coverage degree.

Figure 7 shows the Langmuir plot for the experimental data at 303K, 313K, and 323K, respectively. From the plot of C/θ against C , straight-line graphs were achieved. The results showed that the slopes and R^2 values were almost unity revealing that the adsorption obeyed Langmuir isotherm [24,25]. The Langmuir isotherm parameters are depicted in Table 2.

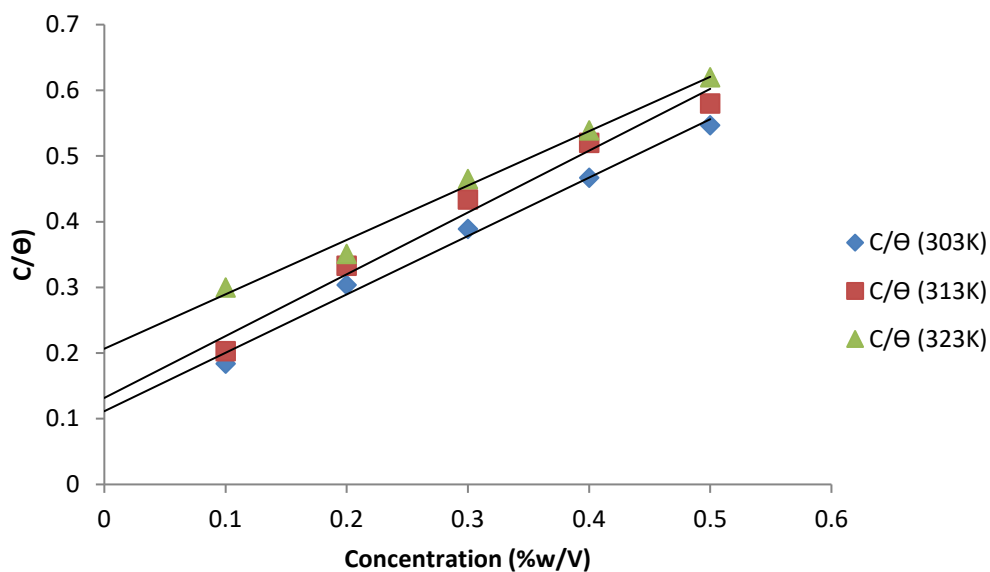


Fig. 7: Plot of C/θ against C at varying temperatures

Table 2: Adsorption Isotherm Parameters

Isotherm	Temp (°C)	$\frac{1}{y}$	Slope	R²	K_{ads}	ΔG_{ads}(kJ/mol)	a	n
Langmuir	30	-	0.889	0.991	9.009	-15.66	-	-
	40	-	0.941	0.980	7.634	-15.74	-	-
	50	-	0.828	0.990	4.854	-15.03	-	-
Freundlich	30	-	0.313	0.986	97.27	-21.65	-	3.19
	40	-	0.426	0.995	99.54	-22.43	-	2.35
	50	-	0.697	0.984	124.45	-23.74	-	1.43
El- Awardy	30	0.754	1.326	0.930	20.51	-17.73	-	-
	40	0.924	1.082	0.915	9.95	-16.43	-	-
	50	0.785	1.274	0.990	9.44	-16.82	-	-
Temkin	30	-	0.539	0.982	94.52	-21.58	-2.14	-
	40	-	0.512	0.966	82.95	-21.95	-2.15	-
	50	-	0.663	0.992	38.83	-20.62	-1.82	-

The resulting slopes are nearly equal to one, demonstrating that the adsorption of the extract was on a single layer of the metal and that there was no interaction between the extract and mild steel [16].

The Temkin Isotherm model was represented by the equation [10,26].

$$\exp(-2a\theta)=KC \quad (7)$$

where the molecules interaction parameter stands for a , Θ = degree of surface coverage, K = equilibrium constant of the adsorption, and concentration of the inhibitor stands for C . θ is related to the C according to the Temkin isotherm.

Rearranging and taking the logarithm of equation 7, equations 8 and 9 were generated as follows;

$$\Theta = \frac{-2.303 \log K}{2a} - \frac{2.303 \log C}{2a} \quad (8)$$

$$\Theta = \frac{-\ln K}{2a} - \frac{\ln C}{2a} \quad (9)$$

$$\text{Slope} = \frac{-2.303}{2a} \quad \text{while Intercept} = \frac{-2.303 \log K}{2a}$$

The plot of surface coverage (Θ) against $\log C$ can be generated using the above Equations.

Figure 8 shows the Temkin isotherm plots for the adsorption of *Solanum macrocarpon* extract on mild steel surface at 303K, 313K, and 323K, respectively.

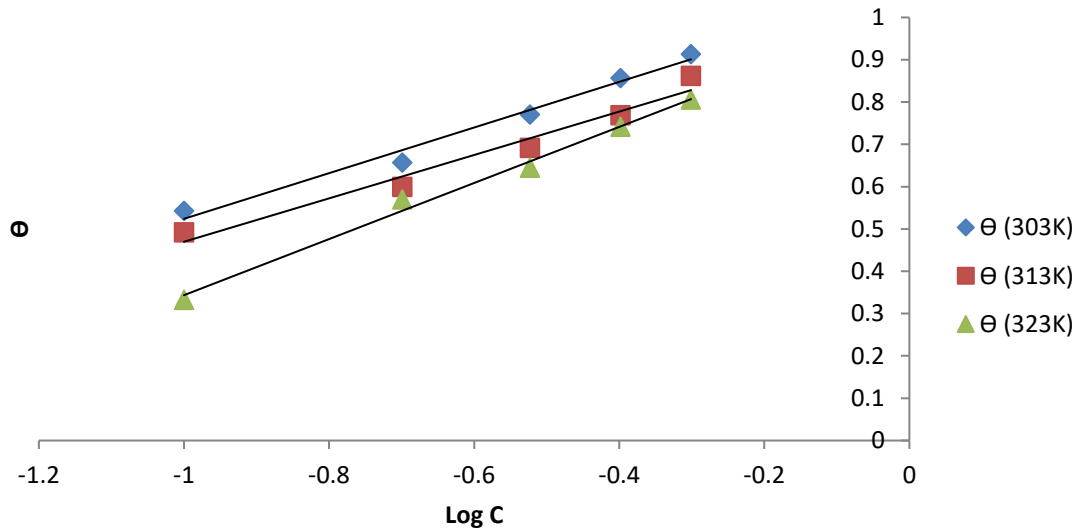


Fig. 8: Plot of Θ versus Log C at different temperatures

Linear plots were derived from the plot of Θ against $\log C$, which showed that the adsorption obeyed Temkin isotherm. The values of correlation coefficient (R^2), slope, intercept, and other Temkin parameters are shown in Table 2. The R^2 values were close to unity, indicating that the experimental data could be modeled by Temkin isotherm. The calculated values of 'a', which is the Temkin/molecules interaction parameter, were all negative, demonstrating the presence of repulsion in the adsorption layer [26].

Freundlich adsorption isotherm was given by the following Equations [17].

$$\frac{x}{m} = KC^{1/n} \quad (10)$$

$$\log \frac{x}{m} = \log K + \frac{1}{n} \log C \quad (11)$$

The fraction $\frac{x}{m}$ represents the inhibition efficiency, where K and n are constants whose values depend upon the adsorbate and adsorbent at a particular temperature (with n values > 1), slope = $\frac{1}{n}$ while Intercept = $\log K$.

The plots of $\log I \%$ versus $\log C$ produced straight line graphs with $R^2 > 0.9$, which obeyed Freundlich adsorption isotherm [23,27]. Figure 9 shows the Freundlich isotherm plots.

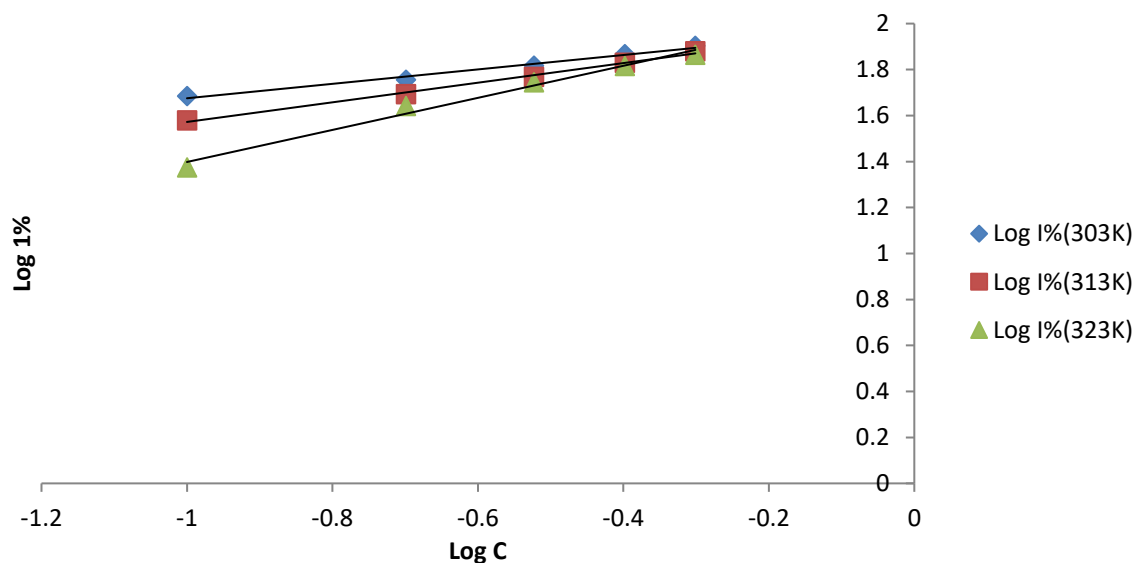


Fig.9: Plot of Log I% against Log C at varying temperatures

The values of other Freundlich adsorption parameters are shown in Table 2. The average slope values were close to the typical value of 0.6, which implied that adsorption fitted into the Freundlich isotherm [28]. The slope values were less than one ($0 < 1/n < 1$), indicating normal adsorption. The value of n , which represents adsorption intensity, was found to lie between one and ten, showing a favourable adsorption process [21,29].

The adsorption process was also studied using the El-Awardy isotherm, which can be represented by the equation [30].

$$\text{Log} \left(\frac{\theta}{1-\theta} \right) = \text{Log } K + y \text{ Log } C \quad (12)$$

Where Θ = degree of surface coverage, K = equilibrium constant, $K_{\text{ads}} = K^{1/y}$, y = the number of molecules at a specific site, $1/y$ = the number of active regions a single inhibitor molecule occupies on a metal surface, C = inhibitor concentration

The plots of $\log \left(\frac{\theta}{1-\theta} \right)$ against $\log C$ produced straight line graphs with $R^2 > 0.9$, which indicated that the adsorption process obeyed El-Awardy isotherm [30]. The El-Awardy adsorption isotherm plot is depicted in Figure 10. The table also shows the El-Awardy adsorption parameters.

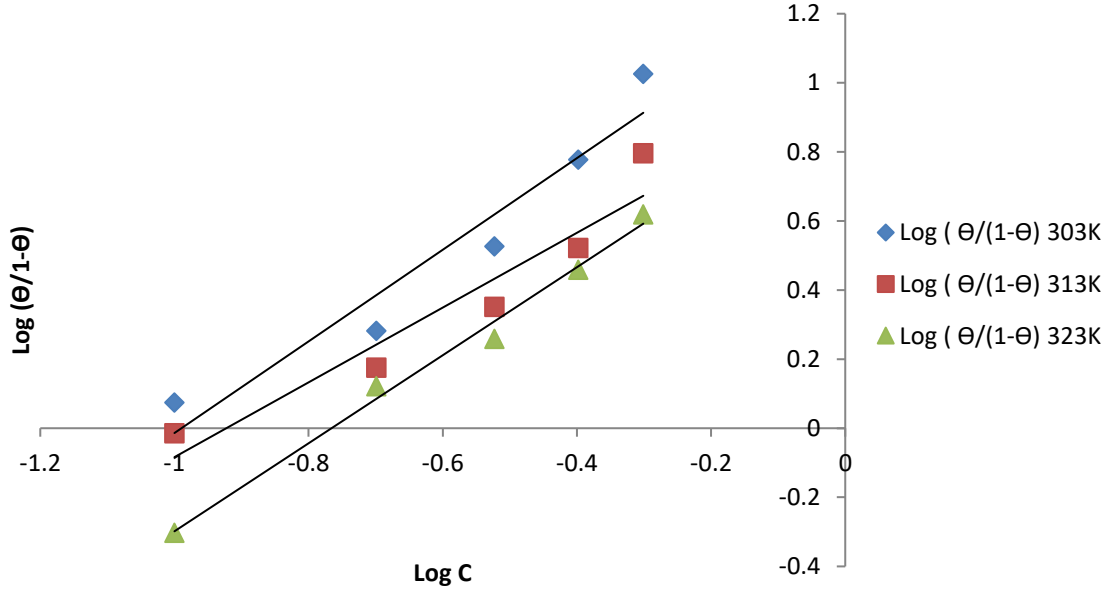


Fig.10: Plot of $\text{Log} \left(\frac{\theta}{1-\theta} \right)$ against $\text{Log} C$ at varying temperatures

The slope values obtained for the experiment at various temperatures studied were found to be approximately unity which showed that only one inhibitor molecule was adsorbed per active adsorption site [18]. Considering that y represents the number of inhibitor molecules at a specific site on the metal surface, the slope values indicated that each adsorption site only received one inhibitor molecule [28]. Furthermore, it has been stated that the number of active sites that one inhibitor molecule occupies on the metal surface is represented by the reciprocal of y ($1/y$) [19]. A particular inhibitor is likely to occupy more than one adsorption site if the values of $1/y$ are larger than unity [21]. The values of $1/y$ were found to be approximately unity which showed

that the inhibitor molecule occupied only one active site of the metal surface, implying that one water molecule was displaced during the inhibition process [17].

The amount of adsorption between the adsorbent and adsorbate is indicated by the constant K_{ads} [31]. The values of K_{ads} calculated from the plots (Table 2) decreased with temperature rise, attributed to the weak interaction between the adsorbing molecules and the mild steel surface, justifying the physisorption mechanism [1].

The Gibbs free energy from the intercept of the adsorption isotherms was represented in Equation 15.

$$\Delta G_{ads} = -2.303RT \log (55.5K) \quad (13)$$

where R = gas constant, T = temperature, K = equilibrium constant, 55.5 = molar heat of adsorption of water. K values obtained from the intercept of the isotherms were utilized in the calculation ΔG_{ads} using Eq. 13, and the result showed in Table 2. The free energy of adsorption ΔG_{ads} results showed negative and lower than -40 kJmol^{-1} required for chemical adsorption, this suggests that the physical adsorption method was followed and the adsorption was spontaneous [11].

4.0 Conclusion

It has been shown in this study that the addition of *Solanum macrocarpon* leaves extract to 2M H_2SO_4 solutions greatly retarded the corrosion rate of mild steel in the acid solutions. The inhibition efficiency of the extract increases exponentially with an increase in concentrations and decreases with an increase in temperature. The inhibition potential of the extract is due to the presence of phytochemical compounds that contain heteroatoms in their functional groups. The

extract adsorption is exothermic and spontaneous, supporting the physical adsorption mechanism by the values of Q_{ads} , ΔG_{ads} , and E_a obtained. The experimental data fit well into Langmuir Isotherm but showed good fits for Temkin, El-Awardy, and Freundlich isotherm. Therefore, this study showed that *Solanum macrocarpon* is a cost-effective green inhibitor and could be utilized in industries to reduce metal corrosion.

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Data availability: All data generated or analyzed during this study are included in this published article

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