Corrosion Inhibition of Cu-Zn-Fe Alloy in Hydrochloric Acid Medium by Crude Ethanol Extracts from Roots-Leaves Synergy of *Solanum melongena*

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Abstract

The corrosion inhibition of Cu-Zn-Fe alloy in hydrochloric acid medium by crude ethanol extracts from roots-leaves synergy of *Solanum melongena* have been studied with chemical methods (mass loss and gasometric methods). At 3.0 grams per litre concentration of the roots-leaves synergy of *Solanum melongena*, it was observed that a 98.8 % inhibition efficiency was recorded as corrosion rate of alloy was decreasing with inhibitor increase. Temperature evaluation on the inhibitor showed 99.2 > 88.4 > 85.6 % as trial was conducted from 303-323 respectively, and in respect to increasing concentration, corrosion rate was found to be 1.718 > 0.013, 0.0192 and 0.247 at 303, 313 and 323 respectively. All these present a good result for the synergistic inhibitor and a proof of its efficiency in controlling the corrosion of Cu-Zn-Fe alloy in hydrochloric acid medium. Inhibition mechanism was deduced from the activation and thermodynamic parameters that govern the process. Adsorption of extract on the Cu-Zn-Fe alloy was found to obey the Langmuir adsorption isotherm. The phenomenon of physical adsorption is proposed from the obtained thermodynamic parameters.

1. Introduction

Corrosion is a natural process since all natural processes tend toward the lowest possible energy states [1-3]. The most common kinds of corrosion result from electrochemical reactions. General corrosion occurs when most or all of the atoms on the same metal surface are oxidized, damaging the entire surface. Most metals are easily

oxidized: they tend to lose electrons to oxygen (and other substances) in the air or in water. The effects of corrosion in our daily lives are both direct, in that corrosion affects the useful service lives of our possessions, and indirect, in that producers and suppliers of goods and services incur corrosion costs, which they pass unto the consumers [2-5]. Some of the effects of corrosion include a significant deterioration of natural and historic monuments as well as increase the risk of catastrophic equipment failures. Corrosion also degrades important infrastructure such as steel-reinforced-highways, electrical towers, parking structures and bridges [4, 5-7]. Its effects can also be seen in plant shutdown, waste of valuable resources, loss or contamination of products, reduction in efficiency, and expensive overdesign. Corrosion damage can be prevented by using various methods such as upgrading materials, blending of production fluids, process control and chemical inhibition [2, 5-7]. The known effects of most synthetic corrosion inhibitors are the motivation for the use of some natural products. Natural products are nontoxic, biodegradable and readily available. Green and eco-friendly inhibitor research have become a wide spread area of corrosion control especially due to their environmental friendly, less expensive and easy availability and accessibility nature [1-3, 7-10]. Their effective and wide spread interest is also centered on the presence of hetero-atoms and dominance of aromatic characteristics, and alkyl groups that act to reduce corrosion through adsorption on metal and alloy surfaces the plant extracts are considered as an incredibly rich source of environmentally acceptable corrosion inhibitors [8-11]. This work is aimed at investigating Corrosion inhibition of Cu-Zn-Fe alloy in hydrochloric acid medium by crude ethanol extracts from roots-leaves synergy of *Solanum melongena*

2. Experimental

2.1 Preparation of ethanol extracts of Solanum melongena

The required roots and leaves of *Solanum melongena* were dried separately in a laboratory oven at a minimal temperature to avoid loss of major organic components of the plant and ground into powder form. Each powdered sample was extracted continually with absolute ethanol in a Soxhlet extractor for 48 hours. The extracts obtained separately were later evaporated of the excess ethanol through a water bath at $60^{o}C$. Ten grams each of the ethanol extracts of leaf and root were weighed and together diluted with 1000 ml 1 M HCl solution then kept for 24 hours to allow for complete dissolution. From the stock solution (10 g/L), inhibitor test solutions were prepared to obtain 1.0 g/L, 1.5 g/L, 2.0 g/L, 2.5 g/L and 3.0 g/L for mass loss and gasometric measurements respectively. The prepared solutions were then used to study the corrosion inhibition abilities of the extract.

2.2 Mass loss measurements

Prepared and weighed 5.0 cm x 0.08 cm Cu-Zn-Fe alloy dimension were considered for the mass loss experiment. These metals were immersed in various concentrations including the blank solution of the synergistic inhibitor using a 100 ml beaker and suspended with a polythene rope and a glass rod. Each of the test specimens were removed daily from the test solution, washed with distilled water, rinsed with ethanol, dried with acetone and re-weighed. Plots of weight loss against exposure time and concentration were generated and corrosion rates obtained from plots. Surface coverage and inhibition efficiency were estimated from equations (1) and (2) respectively:

$$\theta = 1 - \frac{W_1}{W_2} \tag{1}$$

$$\text{IE\%} = \theta \times 100 \tag{2}$$

where θ is the surface coverage, W_1 is the corrosion rate of the blank, W_2 is the corrosion rate of the inhibitor, IE% is the inhibition efficiency.

2.3 Hydrogen evolution measurements

100 mL of the 1 M HCl was introduced into the assembly and the initial volume of the indicator was noted. This was followed with the prepared and weighed 1.20 cm x 0.08 cm x 4.00 cm Cu-Zn-Fe alloy dimension dropped into the blank solution (1 M HCl) and the flask quickly closed. The volume of the hydrogen gas evolved from the corrosion reaction was monitored by volume changes in assembly after every minute for 30 minutes. In another experiment, a set of fresh alloy were immersed in the flask containing the inhibitor solutions (1.0 g/L, 1.5 g/L, 2.0 g/L, 2.5 g/L and 3.0 g/L) and the experiment repeated again. The study was conducted at 303K, 313 and 323 K using a thermostat water bath.

3. Results and Discussion

3.1 Effects of inhibitor concentration on mass loss of Cu-Zn-Fe alloy

Mass loss of metal or alloy is a consequence of the corrosion rate of the alloy due to oxidation at the anodic sites and cathodic hydrogen evolution [6, 9, 11]. The Cu-Zn-Fe alloy was found from Figures 1 and 2 to experience a increase in mass loss with increase in immersion time which could be as a result of increase retention of dissolved alloy particles which serves as bulk impurities [10-13]. The same mass loss was found decreasing with increase concentration implying that the corrosion sites in the alloy were

inhibited by the root-leaves synergy extracts [11-15]. This has been confirmed from Table 1 and Figure 3a-b were surface coverage (θ) and inhibition efficiency (IE%) of Cu-Zn-Fe alloy increased with increase in inhibitor concentrations proving that the corrosion of the metal has been inhibited and a larger fraction of the surface is protected against alkaline attack [13, 16].
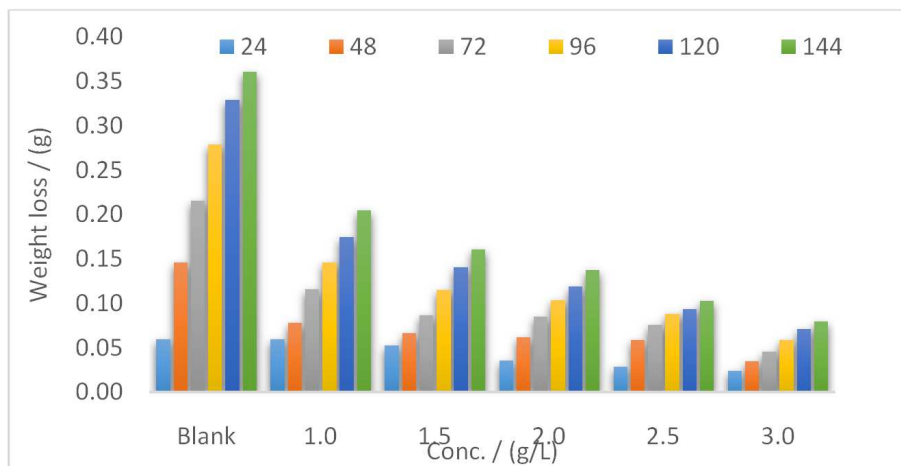

Figure 1. Variation of weight loss with immersion time for Cu-Zn-Fe alloy in the presence and absence of root-leaves synergy extracts of *Solanum melongena* leaves.
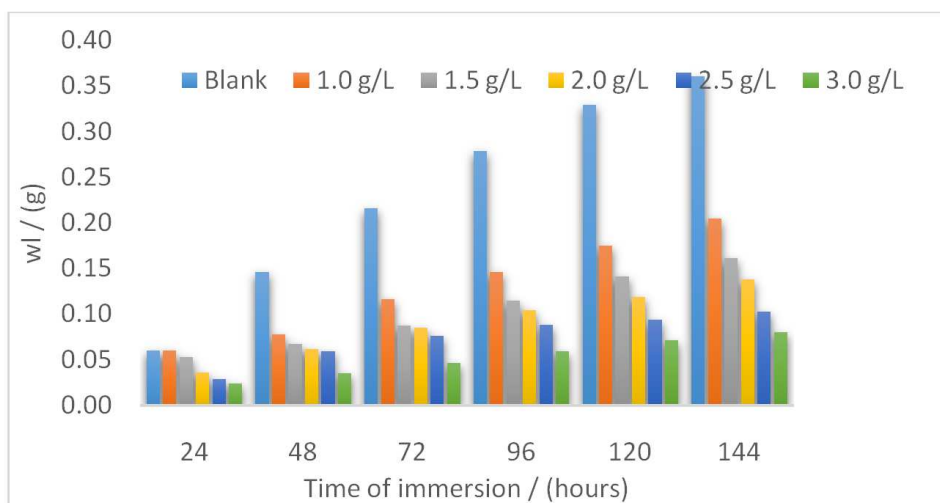

Figure 2. Variation of weight loss with concentration for Cu-Zn-Fe alloy in the presence and absence of root-leaves synergy extracts of *Solanum melongena* leaves.

Table 1. Corrosion rates, surface coverage and inhibition efficiency for Cu-Zn-Fe alloy in the presence and absence of root-leaves synergy extracts of *Solanum melongena* leaves.

System	CR (mg/cm^2/hr)	θ	IE%
(Blank)	1.027	-	-
1.0 g/L	0.237	0.769	76.9
1.5 g/L	0.189	0.816	81.6
2.0 g/L	0.113	0.889	88.9
2.5 g/L	0.096	0.907	90.7
3.0 g/L	0.033	0.988	98.8

CR= Corrosion rate θ = Surface Coverage   IE% = Inhibition efficiency

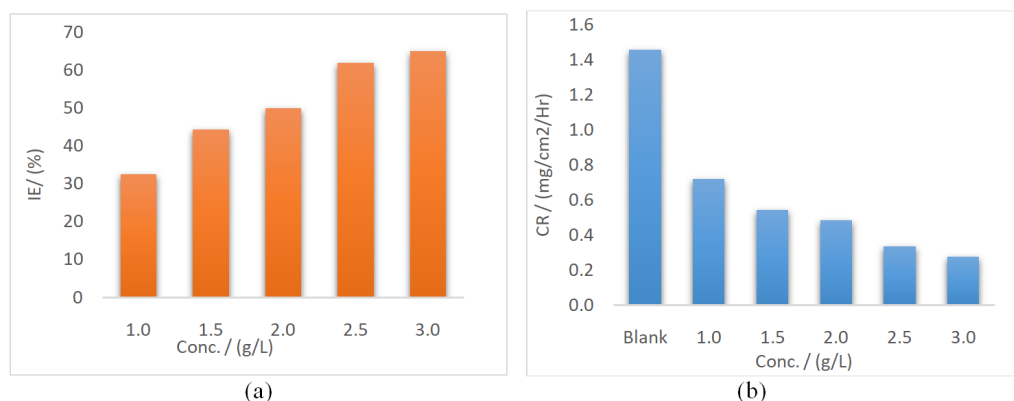(a)   (b)

Figure 3. Variation of (a) IE(%) and (b) CR with Conc. for Cu-Zn-Fe alloy in the presence and absence of root-leaves synergy extracts of *Solanum melongena* leaves.

3.2 Effects of temperature on hydrogen gas evolution in Cu-Zn-Fe alloy inhibition

Table 2 revealed an inhibitor efficiency of 99.2 % > 88.4 % > 85.6 % as test was conducted between 303-323 K respectively, while in respect to increasing concentration, corrosion rate was found to be 1.718 (Blank) > 0.013 (303 K), 0.0192 (313K) and 0.247 (323K). The first scenario informed the gradual desorption of the weakly held inhibitor molecules on the Cu-Zn-Fe alloy surface due to strong increase temperature agitation from 303 to 323 K [12, 15, 16-20]. This decrease in inhibition efficiency with increase

temperature reveals a physical adsorption process [11-16]. The decrease in corrosion rate of Cu-Zn-Fe alloy as concentration increases explained the strong adsorption of the inhibitor molecules even at agitated temperatures [20-22] and all these present a good result for the synergistic inhibitor and a proof of its efficiency in controlling the corrosion of Cu-Zn-Fe alloy in hydrochloric acid medium. This has been summarized on the chart in Figures 4a-c.
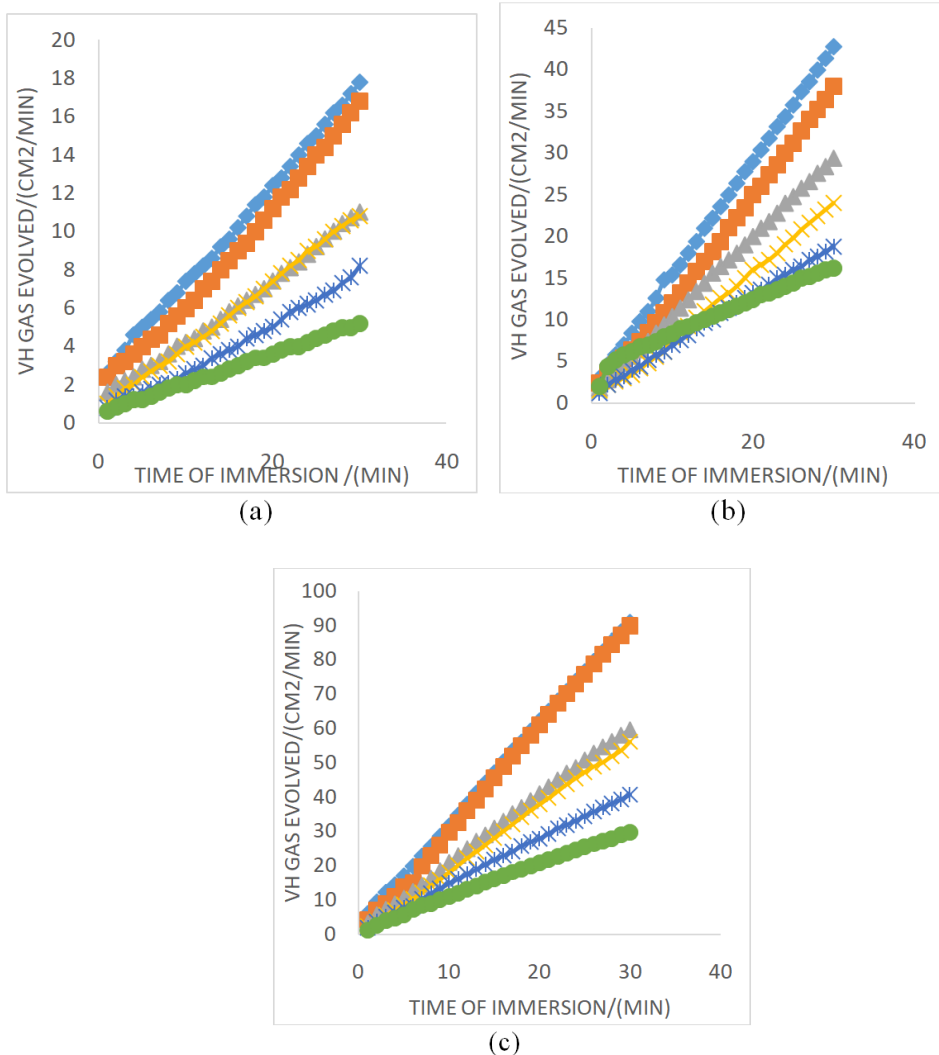

Figure 4. Variation of weight loss with concentration for Cu-Zn-Fe Alloy in the presence and absence of root-leaves synergy extracts of *Solanum melongena* leaves.

Table 2. Corrosion rate, surface coverage and inhibition efficiency for Cu-Zn-Fe alloy in the presence and absence of root-leaves synergy extracts of *Solanum melongena* leaves.

	303K			313K			323K		
Inhibitor Conc.	CR (mg/cm2/min)	θ	IE (%)	CR (mg/cm2/min)	θ	IE (%)	CR (mg/cm2/min)	θ	IE (%)
Blank	1.718	-	-	1.718	-	-	1.718	-	-
1.0 g/L	0.173	0.899	89.9	0.395	0.770	77.0	0.471	0.726	72.6
1.5 g/L	0.099	0.942	94.2	0.375	0.782	78.2	0.471	0.770	77.0
2.0 g/L	0.047	0.973	97.3	0.291	0.831	83.1	0.379	0.779	77.9
2.5 g/L	0.022	0.987	98.7	0.276	0.839	83.9	0.348	0.797	79.7
3.0 g/L	0.013	0.992	99.2	0.192	0.884	88.4	0.247	0.856	85.6

3.3 Thermodynamics

The temperature of the system was varied across the inhibitor concentrations from which the activation energy for the corrosion of Cu-Zn-Fe alloy in 1 M HCl was evaluated using the Arrhenius equation given by equation 3.

$$\ln R_c = \ln A - \frac{E_a}{RT} \tag{3}$$

where R_c is the corrosion rate, E_a is the apparent effective activation energy, R is the general gas constant, and A is the Arrhenius pre-exponential. Calculated values of Activation energy between 303 K and 323 K were obtained from the slope of Figure 5 and presented in Table 3. The values obtained in the inhibitor solution are greater than the value for the blank solution indicating that inhibitor retards the corrosion of Cu-Zn-Fe alloy in 1 M HCl solution [9, 13, 22-24]. Since the activation energy increased with inhibitor concentration, it implies that more energy has to be supplied to the system for the corrosion reaction to take place thus the observed decrease in corrosion rate [14, 22, 24-26]. The values are also consistent with the data expected for the mechanism of physical adsorption (<80 kJmol^{-1}) [17-21]. Enthalpy (ΔH_{ads}), Entropy (ΔS_{ads}) and Heat of adsorption (Q_{ads}) of *Solanum melongena* root-leaves extracts on Cu-Zn-Fe alloy were calculated using equations 4 (transition state equation) and 5 (for heat of adsorption equation),

$$\frac{CR}{T} = \log \frac{R}{Nh} + \frac{\Delta S}{2.303R} - \frac{\Delta H}{2.303RT} \log \tag{4}$$

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

where Q_{ads} is the heat of adsorption, R is the universal gas constant, θ_1 and θ_2 are the degrees of surface coverage of the inhibitors at temperatures T_1 and T_2 respectively. From equation 4, values of $\log(CR/T)$ were plotted against $1/T$ as shown in Figure 6 and from the slop and intercept of the plot, values of enthalpy and entropy of adsorption were calculated as shown in Table 3. From the calculated values of ΔH^* (Table 3), it can be deduced that the adsorption of the inhibitor on Cu-Zn-Fe alloy surface is exothermic and the reaction becomes less exothermic with increase in inhibitor concentration [24-28]. The negative values of Q_{ads} indicated that the degree of surface coverage decrease with rise in temperature, supporting the earlier assumptions of physisorption mechanism for the inhibitor. The negative values for ΔS^* shows the non-spontaneous dissolution of the Cu-Zn-Fe alloy and the increase in its values suggests decrease in disordering in the rate determining step [26-29].

Table 3. Values of activation/thermodynamic parameters for carbon steel in 5 M Na$_2$CO$_3$ in the absence and presence of the plant extracts

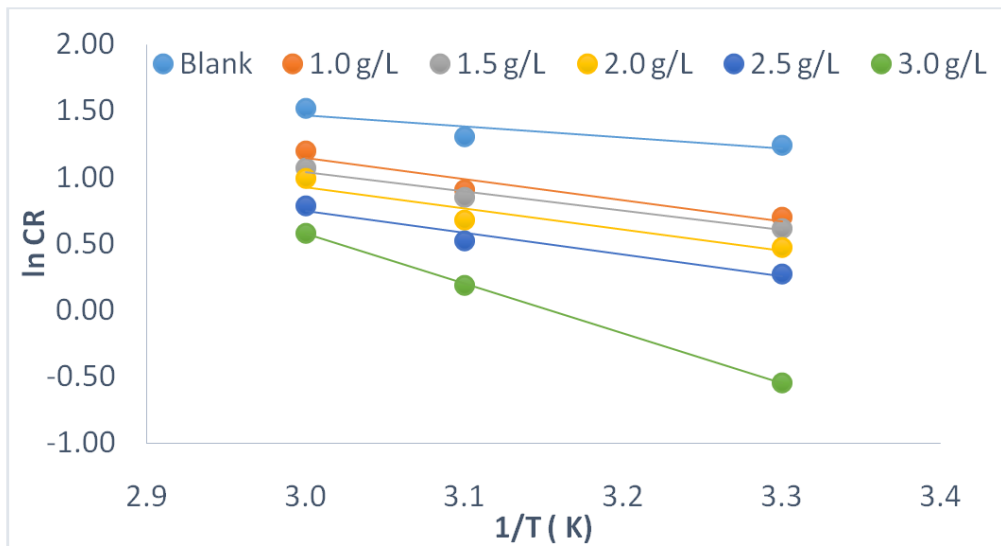| System	Ea (kJ/mol)	ΔH^*_{ads} (kJ/mol)	ΔS^*_{ads} (kJ/mol)	Q_{ads} (kJ/mol)
Blank	6.9	66.9	-76.4	-
1.0 g/L	10.0	74.5	-62.0	-36.10
1.5 g/L	11.8	104.7	-35.9	-43.21
2.0 g/L	11.8	104.7	-27.9	-44.14
2.5 g/L	12.1	123.8	-24.1	-55.80
3.0 g/L	14.1	131.3	-21.4	-72.65

Figure 5. Arrhenius plots for corrosion of Cu-Zn-Fe alloy in the presence and absence of root-leaves synergy extracts of *Solanum melongena* leaves.
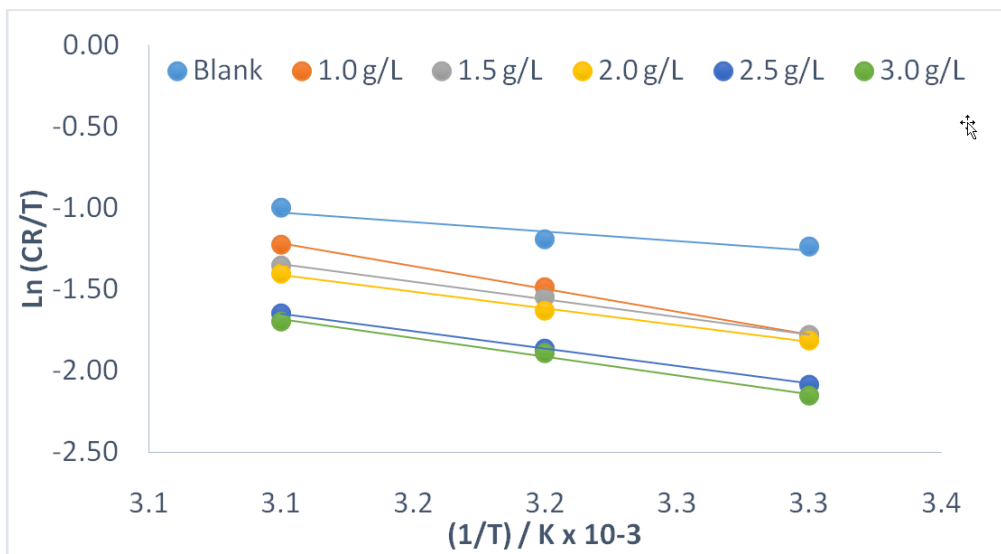

Figure 6. Transition state plots for corrosion of Cu-Zn-Fe alloy in the presence and absence of root-leaves synergy extracts of *Solanum melongena* leaves.

3.4 Adsorption isotherm consideration

The surface coverage (θ) values for different concentrations of the inhibitors in 1 M HCl solutions was evaluated using gasometric data. The data were tested graphically to find a suitable adsorption isotherm to describe the adsorption characteristics of the extracts. A plot of Log (θ/C) against Log C (Figure 7) showed a straight line with regression coefficient of approximately unity (Table 4) indicating that adsorption followed the Langmuir adsorption isotherm [8, 15, 29-31]. The values for the equilibrium adsorption-desorption constant showed decrease with increasing temperature. This is an indication of good inhibition at lower temperatures hence a physical adsorption process just as indicated in Table 2 [22-23, 31-33]. The Gibbs free energy of adsorption values were negative and less than 20 kJ/mol indicating a physical adsorption, inhibitor stability and spontaneity at the forward reaction [31-34].
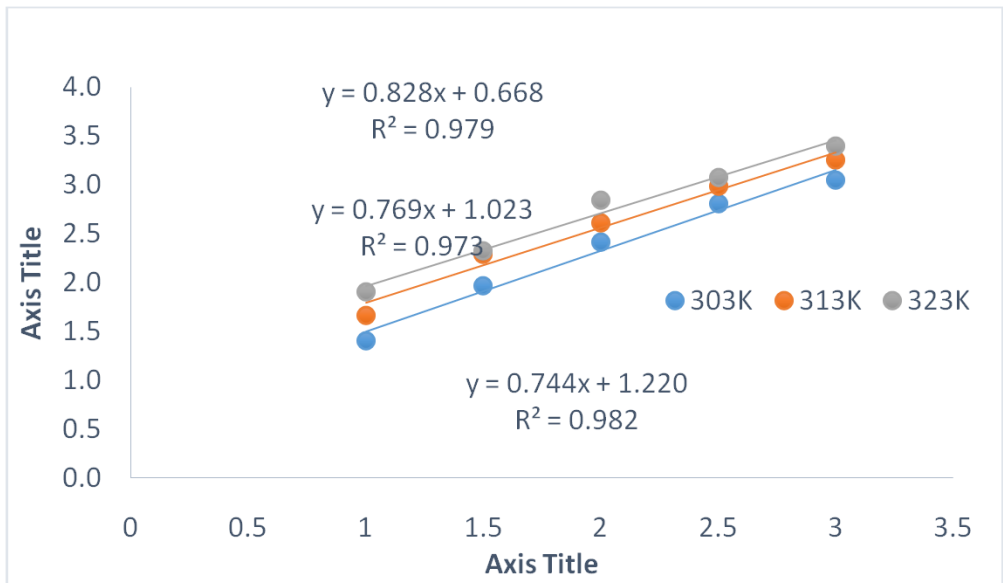

Figure 7. Langmuir adsorption isotherm for corrosion of Cu-Zn-Fe alloy in the presence and absence of root-leaves synergy extracts *of Solanum melongena* leaves.

Table 4. Adsorption parameters for corrosion of Cu-Zn-Fe alloy in the presence and absence of root-leaves synergy extracts of *Solanum melongena* leaves

Temp. (K)	Equil. Const. k (g/L)	Corr. Coef. (R^2)	ΔG^{*}_{ads} (KJ/mol)
303	0.8191	0.9829	-444.22
313	0.9773	0.9737	-318.40
323	1.4959	0.9797	-171.33

Conclusion

1. The percentage inhibition efficiency obtained from this experiment for corrosion inhibition of Cu-Zn-Fe alloy using synergy of root-leaves extracts of *Solanum melongena* leaves showed significant inhibition.

2. Inhibition was by adsorption of molecules of the combined extracts on the surface of the Cu-Zn-Fe alloy there by retarding the anodic dissolution and cathodic hydrogen evolution.

3. Adsorption study reveals that the mechanism of adsorption follows Langmuir isotherm which implies a monolayer adsorption calculated from the correlation coefficient of 0.999 approximate.

4. Thermodynamic study reveals an inhibitor that was physical adsorped, endothermic, stabled, spontaneous and with a very insignificant degree of disorderliness

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