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ORIGINAL ARTICLE

Garcinia kola Leaf Extract as Green Inhibitor for the Corrosion of Aluminium Alloy in Acidic Medium

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ABSTRACT

The effectiveness of Garcinia kola leaf extract in inhibiting aluminium corrosion in 1 M HCl media was investigated. The phytochemical analysis of the Garcinia kola extract showed that it contains phytochemicals including saponin, alkaloid, flavonoid, and tannin. Garcinia kola is a highly effective inhibitor for the corrosion of aluminium in HCl acid environments, and its inhibitory efficiency grows with respect to the quantities of the inhibitor, according to experiments done by weight loss technique. From the study, the corrosion rate increases with increasing temperature and reduces in the presence of plant extracts as the concentration of the inhibitor increases. More so, the effectiveness of Garcinia kola's inhibition was found to diminish with rising temperature and to increase with respect to concentration. The maximum reported inhibitory efficiency was observed at 1.0 M concentration at 318 K, which was 99.03%. This shows that Garcinia kola is a potent organic inhibitor, probably as a result of the inhibitor molecule adhering to the surface of the aluminium alloy.

1 Introduction

Corrosion is the degradation of a material or its qualities as a result of interactions between the material and its surroundings (Adejoro et al., 2015). A metal's propensity to corrode can be affected by its grain structure, its composition as produced during alloying, or the temperature at which a single metal surface developed during manufacturing can deform. It would be more practical to avoid corrosion rather than strive to entirely eradicate it.

Because the environment plays a significant role in corrosion, it is difficult to comprehend (Obot and Obi 2009) (Fragoza et al., 2012). Garcinia kola leaf extract will be utilized in this study to prevent rusting of aluminium alloy. Metal reactivity, the presence of impurities, the presence of oxygen, moisture, gases like sulphur dioxide and carbon dioxide, and the presence of electrolytes are all factors that contribute to corrosion. These issues are addressed via corrosion prevention and retardation (Sastri 2014). Acid solutions are typically used to remove unwanted scale and rust from metals, clean boilers, and heat exchangers, acidize oil wells to recover oil, among other things (Avdeev et al., 2013).

The corrosion of metals can be effectively inhibited by organic compounds that contain heteroatoms with high electron densities, such as phosphorus, sulphur, nitrogen, or oxygen, or those that contain multiple bonds, according to earlier studies (Solmaz et al., 2011). These compounds are also thought of as adsorption centers. (Doner et al., 2013). Alkaloids, tannin, saponin, flavonoids, and phenol are among the phytochemicals that have been found to be abundant in adsorption heteroatoms in Garcinia kola leaf (Ibedu et al., 2018).

In furtherance of our ongoing exploration of green corrosion inhibitors, the current work reports on the inhibitory impact of

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Garcinia kola leaf extract for aluminium in a hydrochloric acid environment utilizing chemical and electrochemical techniques.

According to Uguru et al. (2019), the inhibitory efficacy of Garcinia kola increased from 67.21 to 88.19% with a rise in temperature in the HCl acidic medium. The investigation also revealed that the corrosion rate dropped from 0.37 (0% inhibitor concentration) to 0.03 (40%) and that an increase in temperature improved inhibition effectiveness. It might be said that bitter kola leaves are effective at inhibiting corrosion in 1 M HCl.

The slopes of the linear parts of the hydrogen evolution versus time plots and the associated values for the various test solutions were used to estimate the rates of corrosion of the carbon steel coupons. The findings demonstrate that for all systems, corrosion rates rose with increasing temperature and fell in the presence of Garcinia kola extracts (Sanmugapiya et al., 2023). Moreover, it was tested, how temperature (303 and 333 K) affected the corrosion behavior of mild steel when Garcinia kola extract was present.

The findings demonstrate that as the temperature rose, both the lack and presence of the extract had higher rates of corrosion. The effectiveness of inhibition often declines as temperature rises. Due to increased solution agitation brought on by higher rates of H_2 gas evolution, the adsorption-desorption equilibrium may have shifted in favor of the desorption of the adsorbed inhibitor. Moreover, the capacity of the inhibitor to be adsorbed on the metal surface may be hampered by the roughening of the metal surface brought on by accelerated corrosion. The physical adsorption of organic materials on the corroding mild steel surface is suggested by the decrease in inhibition efficiency with temperature rise (Nkem et al., 2022).

Metal parts constitute a majority of automobiles and airplanes, and the deterioration of these parts due to corrosion constitutes a major problem globally. The search for anti-corrosive agents of natural origin is gaining attention due to their lower toxicity. Furthermore, these natural compounds are cheap, biodegradable, and readily available, even in developing countries. This has warranted the need for this study, exploring the potential of this plant extract as anti-corrosive agents.

2 Experimental Section

2.1 Aqueous extraction

Cold maceration was used to extract 50 g of Garcinia kola leaves with 450 ml of ethanol. The ethanol was then evaporated using a water bath at a constant temperature of 45 °C, filtered first through muslin cloth and then through Whatmann No. 4 filter paper, and finally transferred to a sterile sample vial.

2.2 Phytochemical testing

On a sheet of aluminium alloy (AA4007) with the following compositions: Mn (1.2%), Si (1.4%), Ni (0.3%), Cr (0.1%), Fe (0.7%), and Al (96.3%), corrosion experiments were conducted. Aluminium was manually cut, had a hole drilled in one end for free suspension, and had a number punched into it before the corrosion process. After that, each coupon's surface was smoothed out using 220, 800, and 1200 emery sheets. The coupons underwent additional acetone degreasing, were rinsed with distilled water to remove debris, and were then dried in warm air (Ajike et al., 2023).

The coupons were rinsed three times: once with nitric acid to stop the reaction, once with distilled water to get rid of the inhibitor solution, and once more with ethanol and acetone to dry them. The mass loss was calculated using an electronic analytical balance (metlar terado) with a sensitivity of 0.001g to compare the weight of the specimens before and after immersion. To ensure the validity of the findings, the tests were repeated, and the mean value of the mass loss is provided.

Corrosion rate was calculated from mass loss measurements and represented in $mgcm^{-2}hr^{-1}$ for the samples (Khadom et al., 2015). The following formulae were used to calculate the inhibitory efficiency (%) based on weight loss:

$$\Delta w = (w_1 - w_2) \times 1000 \text{ (mg)} \qquad 1)$$

$$C.\,R. = K\Delta \mathrm{w}/A.P.t \qquad 2)$$

$$\text{Inhibition efficiency } (\eta\ \%) = (W_B - W_1)/W_B \times 100 \qquad 3)$$

Where C.R.= corrosion rate, Δw = change in weight (mg), A = cross sectional area of aluminium (cm^2) P = density of aluminium, (g/cm^3), t = time of study (hr), w_1 and w_2 are the initial and final weights of aluminium coupon.

The degree of surface coverage (θ) was calculated from the weight loss measurements using Equation 4:

$$\text{Surface coverage } (\theta) = (W_B - W_1)/W_B \qquad 4)$$

Where W_B and W_1 are the weight loss in the absence and presence of the inhibitor.

In order to evaluate weight loss at 303 K and 318 K, the impact of temperature on the inhibitor's ability to inhibit was investigated.

The Langmuir adsorption isotherm was determined by using Equation 5:

$$C/\theta = 1/K_{ads} + C \qquad 5)$$

Where C is the Garcinia kola concentration, is the surface coverage, and K_{ads} is the equilibrium adsorptive constant (Iroha and Hamilton 2018).

The adsorption free energy (ΔG^0_{ads}) is related to K_{ads} by Equation 6:

$$\Delta G^0_{ads} = -\,RT \ln(K_{ads} \times 55.5) \qquad 6)$$

Where T is the absolute temperature and R is the gas constant (8.314 kJ/mol). The amount of water in solution, expressed in mol/L, is 55.5. At two distinct concentrations of Garcinia kola extracts, 303 K and 318 K, the adsorption parameters from Langmuir adsorption isotherms are computed.

2.3 Studies in Thermodynamics

Knowing the thermodynamic factors that control the adsorption process allows one to predict the manner of adsorption of inhibitors, which can be either physisorption or chemisorption. A variant of the Arrhenius equation was used to compute the activation energies E_a for the corrosion process in the absence and presence of leaf extracts of Garcinia kola:

$$\ln W = \ln A + (-E_a)/2.303RT \qquad 7)$$

W represents the corrosion rate, A is the frequency, factor R is the gas constant and E_a is the activation energy which can be obtained when $\ln W$ is plotted against the inverse of absolute temperature $1/T$ (Iroha and Hamilton 2018). Activation energy is given as:

$$E_a = -\text{ slope} \times 2.303\text{R} \qquad 8)$$

Where R = 8.314 kJ/mol is the real gas constant.

2.4 Potentiodynamic polarization studies

In the potential range between 250 mV from the open circuit potential, potentiodynamic polarization curves were captured (E_{oc}). By automatically varying the electrode potential from -1.850 V to -1.350 V at a scan rate of 25 mVs^{-1}, the measurements were taken. To determine corrosion current densities, the linear Tafel segments of the anodic and cathodic curves were extrapolated to the corrosion potential (E_{corr}) (I_{corr}). The software from EC Lab was used for this. The Equation was used to compute the inhibitory efficiency.

$$\eta\text{PDP } \% = (I^*_{corr} - I_{corr})/I^*_{corr} \times 100 \qquad 9)$$

Where I^*_{corr} and I_{corr} are the corrosion current densities in the absence and presence of the inhibitor, respectively.

2.5 Electrochemical Impedance Spectroscopy

Regarding electrochemical systems, impedance analysis is essential (Wang et al., 2019). For an Al electrode submerged in 1 M HCl in the unprotected media and the protected medium with 100 mg/L and 1000 mg/L, respectively, Nyquist plots were produced. The following factors are taken into consideration in this study: R_{ct}, double layer capacitance (cdl), and (IE%). The main factors derived from the examination of the Nyquist spectra are the double layer capacitance (cdl) and the resistance of charge transfer (R_{ct}), which are each described (Wang et al., 2019) as:

$$\text{Cdl} = 1/(2\pi f_{max} \text{ R}_{ct}) \qquad 10)$$

Where fmax is the rate at which the imaginary part of the impedance is elevated-Zim (max).

3 Results and Discussions

3.1 Phytochemical analysis

The extracts were shown to be high in flavonoids (+++), phenolics (++), alkaloids (+++), saponins (+), and tannins (++) in Table 1. These bioactive components' anti-inflammatory and anti-oxidant properties supported Garcinia kola extracts' anti-corrosion abilities. Dry leaves are the main source of polyphenolic compounds such flavonoids and phenolic acids. Flavonoids with a benzo-pyrone ring structure occur naturally in plant-based materials as a reaction to microbial illness (Kumar and Pandey 2013).

The group of compounds known as phenols is made up of hydroxycinnamic and hydroxybenzoic acids, both of which are found in sufficient amounts in the leaves of dry plants. These phytochemicals' presence suggests that the Garcinia kola leaf extract is an effective organic corrosion inhibitor (Prakash et al., 2007).

Table 1: Phytochemicals found in abundance, mild and in trace in the extracts of Garcinia kola

Phytochemicals	Garcinia kola
Saponin	+
Alkaloids	+++
Tannins	++
Flavonoid	+++
Phenol	++
Reducing Sugar	+

-Absent, +Mild, ++moderate, +++ Abundance

3.2 Corrosion rate

When the concentration of the inhibitor (Garcinia kola) increases, Figure 1 shows that the corrosion rate reduces. Both for 303 K and 318 K, the maximum corrosion rate was seen at the blank specimen. This is explained by the inhibitor molecules' adsorption on the aluminium alloy surface (Onukwuli et al., 2018), which prevents corrosion on the metal surface. Moreover, the corrosion rate rises with temperature, according to Figure 1, the greatest rates were 127.09 mm/yr at 318 K and 67.76 mm/yr at 303 K for the blank specimen, both of which are consistent with the findings of (Ikeuba et al., 2013).

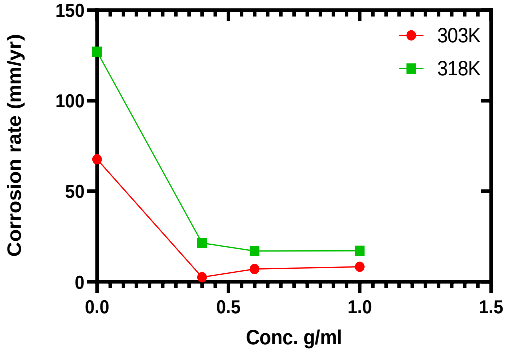

Figure 1: Variation of corrosion rate with Garcinia kola for aluminium in 1.0 M HCl at different temperature

3.3 Inhibition efficiency

Figure 2 shows that temperature variation has an impact on the Garcinia kola extract's ability to prevent cell growth. The composition and thickness of the Garcinia kola film adsorbed on aluminium alloy are influenced by temperature. The efficacy of the inhibition decreases with temperature, which suggests that the extract is physically adsorbed on aluminium. This might occur as a result of inhibitor molecules desorbing from the aluminium surface at a higher temperature (Chaubey et al., 2015). As the organic inhibitor concentration rises, the effectiveness of inhibition increases, as shown in Figure 2. The lowest inhibition effectiveness is 88.35% at the lowest concentration and at 318 K, which is also where the highest inhibition efficiency of 99.09% is found.

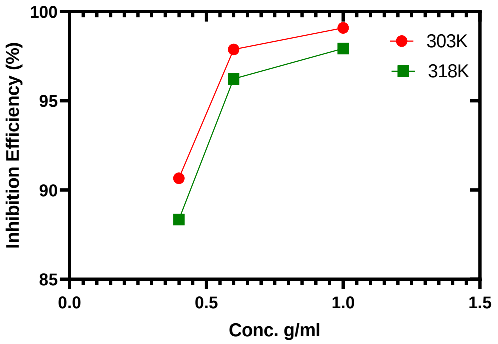

Figure 2: Inhibition efficiency of Garcinia kola ethanol extract in 1 M HCl for aluminium alloy

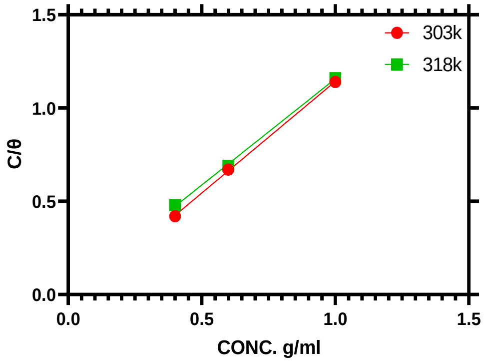

Figure 3: Langmuir isotherms for adsorption of Garcinia kola on aluminium in 1.0 M HCl for 2hrs

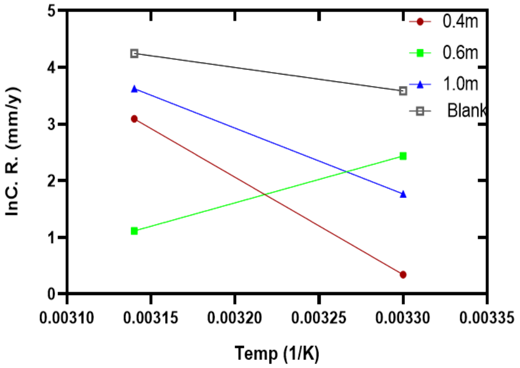

Figure 4: Plot of lnCR versus 1/T for Aluminium corrosion in 1 M HCl (Blank) and inhibited solutions at various concentrations of Garcinia kola for 2hrs

3.4 *Thermodynamic isotherm*

3.4.1 *Isotherm of Langmuir*

Figure 3 below displays the Langmuir plot for Garcinia kola at 303 K and 318 K, respectively. The image clearly shows that the Gibb's free energy of adsorption (G_{ads}), which is connected to the adsorption constant (K_{ads}), Garcinia kola yields a straight line, and all of the regression coefficients (R^2) for 303 K and 318 K are very close to unity (one), indicating that an organic inhibitor molecule has adhered to the surface of aluminium. The values of the slopes from 303 K and 318 K in Table 2 are quite close to unity. The slopes' departure from unity is frequently seen as evidence that the adsorption species have essentially occupied a typical adsorption site at the metal (Umoren et al., 2016).

Table 2: Calculated values of Langmuir isotherm adsorption parameter for 1.0 M HCl Garcinia kola for a) 303 K b) 318 K

(a)	**303 K**			
Time (hr)	**R^2**	**Slope**	**K_{ads}**	**ΔG_{ads} (J/mol)**
2	0.9115	1.211	43.745	-19.6361
(b)	**318 K**			
Time (hr)	**R^2**	**Slope**	**K_{ads}**	**ΔG_{ads} (J/mol)**
2	0.9095	2.804	1.341	-11.3945

The Gibb's free energy values are negative, indicating the spontaneous adsorption of Garcinia kola on the aluminium surface, as evidenced by the values of Gads computed from the Langmuir parameters for 303 K and 318 K of Garcinia kola in Table 2 using equation (6) (Nayem et al., 2020).

The Gibb's free energy values in Table 2(a & b) are less than -20 kJ/mol at 303 K and 318 K, showing that the adsorption of Garcinia kola in an acidic media is physical in nature. Electrostatic interactions between charged molecules and molecules near metallic surfaces cause the inhibition to take place (Rajendral and Karthikeyan 2012).

3.4.2 *Activation energy (E_a)*

Table 3 lists the activation energy values for *Garcinia kola*. The plots as depicted in Figure 4 were used to determine the values. Equation (8) was used to determine the activation energy from the slopes of the plots. The impact of inhibitor concentrations and corrodant concentrations were considered. The activation energy values of the sample containing the organic inhibitor (*Garcinia kola*) were found to be greater than those of the blank sample. This suggests that the inhibitor increases activation energy while lowering reaction rate.

3.5 *Potentiodynamic polarization*

The polarization curves in the presence of the Garcinia kola inhibitor changed towards the more negative potentials in comparison to the blank system, as can be observed in Figure

5, showing the inhibitor's more predominant cathodic inhibitory actions. When the difference in the E_{corr} value between the inhibited and the blank system is more than 85 mV, a chemical may only be classified as either an anodic or cathodic inhibitor (Chokor et al., 2022).

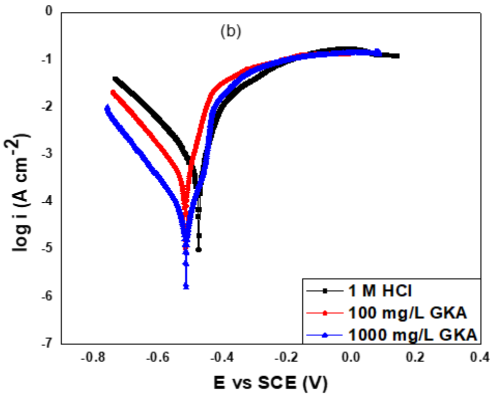

Figure 5: Potentiodynamic polarization plots of aluminium in the presence of Garcinia kola in 1 M HCl environment

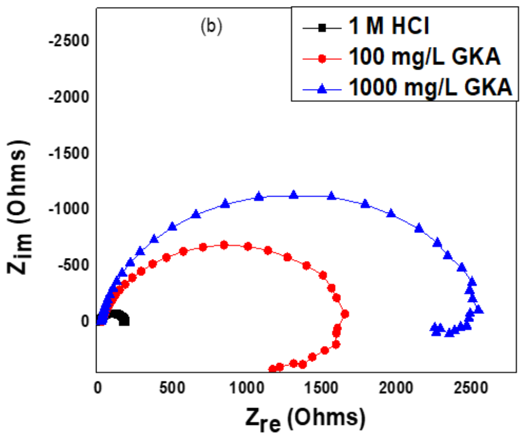

Figure 6: Electrochemical impedance spectra plot of Aluminium in the presence of Garcinia kola in 1 M HCl environment

As the amplitude of the E_{corr} changes from the Blank, is larger than 85 mV and is shown in Table 4's E_{corr} values for Garcinia kola, the extract can be categorized as either anodic or cathodic. With an increase in inhibitor concentration, the corrosion current density, i_{corr} (A/cm^2) values drop.

The obtained inhibitor effectiveness rises with increasing inhibitor concentrations, peaking at 91.5% at 1000 mg/L, which is the opposite of the I_{corr} trend as indicated in Table 4. They show that as the concentration of the inhibitor rises, so does its potency.

Also, it implies that as the concentration of Garcinia kola increases, the rate at which aluminium metal corrodes in 1 M HCl decreases (Nnanna et al., 2010).

3.6 Electrochemical impedance spectroscopy measurement

To learn more about the electrochemical activities that take place at the corroding steel's surface, both with and without the addition of *Garcinia kola*, EIS measurements were conducted. Figure 6 shows the impedance spectra of aluminium metal for *Garcinia kola* in 1 M HCl solution both with and without concentrations of the inhibitor. The presence of a charge-transfer corrosion process that is unaffected by the addition of *Garcinia kola* may be seen in the Nyquist plots' depressed capacitive loop (semicircle).

However, the semicircle's diameter grew as Garcinia kola concentrations raised, indicating that the supplement improved the resistance to charge transfer at the aluminium alloy/1MHCl contact and left a *Garcinia kola* protective coating on the aluminium surface (Lucky and Nkem 2023).

The EIS spectra are also imperfect semicircles that exhibit frequency scattering as a result of the solid electrode's roughness and other inhomogeneities (Scendo and Uznanska 2011). It is clear from Figure 6 that when the concentration of *Garcinia kola* in the 1 M HCl environment rises, so does the semicircle's diameter.

Due to the inhibitor's adsorption on the aluminium alloy surface, this observed increase in diameter denotes a rise in *Garcinia kola*'s capacity to protect against corrosion (Nnanna and Nkem 2020).

Table 3: Activation energy value of Garcinia kola in 1.0 M HCl for aluminium alloy

Time (hr)		**Activation Energy (kJ/mol)**		
	Blank	0.4 m	0.6 m	1.0 m
2	79.35	328.85	158.21	222.36

Table 4: Electrochemical parameters for aluminium alloy in 1M HCl in the absence and presence of Garcinia kola

System	**E_{corr} (mV vs SCE)**	**I_{corr} (μA/cm^2)**	***IE* (%)**	**R_{ct} (Ωcm^2)**	***N***	**R_{LI} (Ωcm^2)**	**I.E (%)**
1 M HCl	- 445.2	218.6		199.7	0.88		
100 mg/L	- 546.3	26.4	87.9	1361.6	0.89	232.7	85.3
1000 mg/L	- 558.9	12.2	94.4	2363.2	0.89	333.1	91.5

4 Conclusion

In 1 M HCl, Garcinia kola effectively inhibited the corrosion of aluminium alloy. The effectiveness of 99.09% inhibition was achieved. The weight loss experiment's findings and those from the EIS and PDP experiments were in excellent agreement. Weight loss measurement results showed higher levels of inhibitory efficiency than electrochemical measures, it was discovered (EIS and PDP). This is explained by the fact that, in contrast to EIS and PDP, weight loss experiment provides average corrosion rates. The experimental data also matched the Langmuir isotherm closely. In conclusion, it is found that Garcinia kola is an effective, environmentally friendly, and biodegradable organic inhibitor.

Declaration of Competing Interest

Authors have declared that no competing interests and funding interests exist. This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

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