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Phytochemical Screening and Corrosion Inhibition of the Ethanolic Leave Extracts of *Gardenia aqualla* Stapf & Hutch In 1M H_2SO_4 Acid Solution

Opara Chinonso Ivan[1], Louis Hitler[2, 6, *], Japari Joseph[1], Oyebanji Oyetola[3, 4], Akakuru Ozioma Udochukwu[2], Tonny Nyong'a Maraga[5], Pigweh Amos Isa[1]

[1]Department of Chemistry, Modibbo Adama University of Technology, Yola, Nigeria
[2]Department of Pure and Applied Chemistry, University of Calabar, Calabar, Nigeria
[3]Department of Botany, University of Lagos, Lagos, Nigeria
[4]Kunming Institute of Botany, University of Chinese Academy of Sciences, Kunming, China
[5]Department of Botany, Jomo Kenyatta University of Agriculture and Technology, Nairobi, Kenya
[6]CAS Key Laboratory for Nanosystem and Hierarchical Fabrication, CAS Centre For Excellence in Nanoscience, National Centre for Nanoscience and Technology, University of Chinese Academy of Sciences, Beijing, China

Email address:
louismuzong@gmail.com (L. Hitler)
[*]Corresponding author

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Abstract: The main objective of this study is to investigate the corrosion inhibition properties of *Gardenia aqualla* leaf extract on mild steel using weight determination loss method. The corrosion of mild steel was investigated at different concentrations of *Gardenia aqualla* at temperatures of 303, 313 and 323 K. *G. aqualla* inhibit corrosion rate of mild steel from 62.4 mm/yr to 6.5 mm/yr, 91.6 mm/yr to 16.6 mm/yr and 113.8 mm/yr to 28.4 mm/yr at 303, 313, and 323 K respectively. The optimum obtained yield in the inhibition efficiency is from 63.8% to 89.5%, 52.5% to 81.9% and 39% to 75% for the various temperatures respectively after an interval of 3 days being in contact in aqueous solution. Langmuir isotherm confirmed that physisorption occurred and it was found that inhibition occurred through adsorption of the constituents present in the leaf of *G. aqualla* obtained by phytochemical screening.

Keywords: Mild Steel, Corrosion Inhibition, *Gardenia aqualla*, Adsorption Mechanism

1. Introduction

An impending phenomenon that is widely experienced in our society, which affects the economy and the society is called corrosion. Its consequences arises when it damages the exterior surface of metals and most times leading to metallic part of an equipment's or tools not working appropriately as expected. It is not only limited to industries or factories, but this happens in almost all environments [1], [2].

The corrosion phenomenon can be attributed to taking place in a number of processes. First, there is a reduction of the mass and thickness of the metal as a consequence of an attack on its surface. Furthermore, in the place of a whole surface attack, certain sites in the area of the metal could be affected, forming the accustomed localized corrosion. Additionally, it also takes place along grain boundaries or other spots on the metal that are weak because of a change in resistance to corrosive menace.

A substance known as an inhibitor tends to stop a chemical reaction or process that reduces the activity of reactants or catalysts on the surface of the mild steel. It enhances the formation of an oxide as a protective film through the effect of oxidation, hence a barrier is created between the metal surface and the selectively adsorbed species, thereby inhibiting corrosion and retarding the process of corrosion or preventing the corrosive agents on the surface of mild steel [3]. Some organic compounds have been studied to know their effective potentials as corrosion inhibitors [4-6].

Most of these studies reveal that the optimum yield in the values of inhibition efficiency is due to the fact that virtually all organic molecules containing heteroatoms (chemical synthetic inhibitors) such as nitrogen, sulphur, phosphorous and oxygen are duly responsible for it [7]. Huge successes have been achieved by the use of these synthetic chemical inhibitors but they pose some risks to human life and its environment. These dangers include; environmental pollution, lack of availability and high cost [8]. It was paramount that these risks ought to be managed, thus compelling scientists to search for their replacement.

Gardenia aqualla Stapf & Hutch. is a flowering plant belonging to the family Rubiaceae and it is found in the tropical and subtropical districts of Africa, including Nigeria as recorded in early studies [9]. *G. aqualla* is a savanna shrub, up to 9 ft. high. Its flowers are white, turning golden-yellow later in the day and fragrant. The fruit of *G. aqualla* are oblong and woody [9]. *G. aqualla* is locally called 'Gaude' in Hausa language, Nigeria [10] and it has been reported to be utilized for ear treatments, oral treatments and used as farming, forestry, hunting and fishing apparatus [11].

Studies have shown that the phytochemistry of the methanolic extract of stem bark of *G. aqualla* revealed the presence of steroid, carbohydrate, anthraquinones, saponins, triterpenes, tannins, cardiac glycoside and flavonoids. The antimicrobial activity of this plant indicates that the plant can be used as traditional medicine [12-14]. Hence, the leaves, roots, fruits and stem bark is used to treat leprosy, oral infection, ear infection and bowel disorder respectively in Northern Nigeria [15].

Researchers in the past have studied the inhibition efficiency and corrosion rate reduction of different plants and the parts used as inhibitors for retarding the corrosive processes. These include black pepper extracts, leaves, seeds and roots extract of *Azadirachta indica* seeds *Foeniculum vulgare* [15-18].

In this present study, an attempt was made to examine the ethanolic extracts of *G. aqualla* leaves as a potential inhibitor of corrosion for mild steel in 1 M H_2SO_4 medium using gravimetric technique along with some statistical model to verify efficiency of the inhibitor developed.

2. Materials and Method

2.1. Source of Plant Material

Leaves of *Gardenia aqualla* Stapf & Hutch. (Figure 1) was collected from Girei local Government Area, Adamawa State, North-East, Nigeria, geographically located on Latitude 9.4265°N and Longitude 12.4809°E, in February, 2017. The plant material was taxonomically identified at the Lagos University of Herbarium (LUH), University of Lagos, Nigeria [19].

Figure 1. *Gardenia aqualla Stapf & Hutch.*

2.2. Preparation of Gardenia aqualla Extract

The leaves were shade dried for 3 days, after which the surface area of the leaves was increased by grinding it with an electric blender. The powdered leaves were soaked in ethanol which was continually stirred with the aid of a magnetic stirrer for 24 hours [5]. The dried alcoholic extracts were prepared by evaporating the filtrate, hence, used for the preparation of inhibitor test solutions in the concentrations of 0.2 g/100ml, 0.4 g/100ml, 0.6 g/100ml, 0.8 g/100ml solution of 1 M H_2SO_4.

2.3. Mild Steel Preparation

The material used for this work that is mild steel, was composed of the following elements namely; C = 0.078, Si = 0.05, P = 0.99, Cu = 0.02, and Fe = 98.86. The specimens were of average dimensions 1.2 cm x 0.72 cm x 0.77 cm which was used for the gravimetric study. The exterior of the specimens were scraped using sand paper, washed thoroughly with acetone and rinsed with distilled water, which was then placed in a desiccator before immersing them into their respective solutions [5].

2.4. Gravimetric Technique

The weighed mild steel specimens, were of identical sizes, and were then suspended in 100 mL test solutions of different concentrations at various temperatures for an interval of 3 days. At every three days interval, the specimens were taken out, washed with distilled water, rinsed with ethanol, dried manually, and weighed again to obtain its new mass. The experiment was carried out in duplicate and the average value stated. The weight-loss was calculated after each measurement. From these data, the various values of the inhibition efficiency (I%), Corrosion rate (Cr) and Surface coverage (θ) was calculated using the following mathematical relationships:

$$I (\%) = \left(\frac{W_1 - W_2}{W_1}\right) \times 100 \quad (1)$$

$$C_r = \frac{K \Delta W}{\rho A t} \quad (2)$$

$$\Theta = \frac{W_1 - W_2}{W_1} \quad (3)$$

where, I (%) is efficiency of inhibitor, W_1 = weight loss of mild steel in the absence of inhibitor, W_2 = weight loss of mild steel in the presence of inhibitor, C_r = corrosion rate in mmy^{-1}, W = weight loss of the mild steel in mg, A = total area of the mild steel sample in cm^2, t = time of exposure of the metal sample in test solution in hours, ρ = density of mild steel (i.e. 7.86 gcm^{-3}), θ = surface coverage of inhibitor on mild steel surface, and K=87.6.

3. Results and Discussion

3.1. Natural Compounds Obtained from the Phytochemical Screening

Table 1 shows the result of the constituents of *Gardenia aqualla* which indicates the presence of saponins, tannins, flavonoids, alkaloids, and steroids while Terpenoids and Glycosides were absent. These natural products present in the extracts serve as a barrier at the surface of mild steel through the mechanism of adsorption, thereby inhibiting or retarding the corrosion process. Though, the blocking of the surface cannot be attributed to a specific or group of constituent (s). Also, the summary of average weight loss (mg), corrosion rate (mm/yr), inhibition efficiency (%), surface coverage (θ) and concentration/surface coverage are shown in table 2.

***Table 1.** Phytochemical screening of the Gardenia aqualla leaves.*

Compounds	Status
Alkaloids	+
Flavonoids	+
Glycosides	-
Phenols	+
Saponins	+
Steroids	+
Tannins	+
Terpenoids	-

Key: Present (+), Absent (-).

3.2. Weight Loss Studies

Results from the gravimetric study such as corrosion rates and efficiency of inhibition at different temperatures and concentrations of *Gardenia aqualla* in 1.0 M H_2SO_4 after 3 days interval is presented in Figures 2 to 4. From these results, it can be comprehended that the rate of corrosion reduced significantly on adding the corrosion inhibitor and then reduced progressively with an increase in inhibitor concentration while inhibition efficiency also increased with an increase in the concentration of the inhibitor until its maximum value of 89.5% was achieved [20], [21] [6].

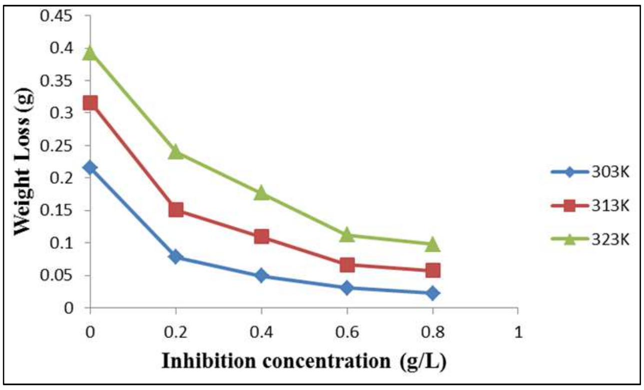

***Figure 2.** Weight loss of Mild steel with and without the Gardenia aqualla extract.*

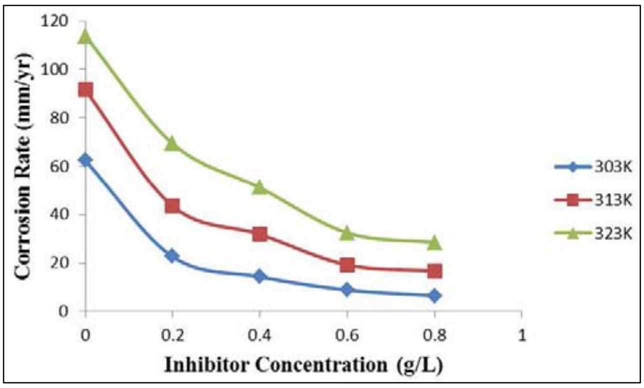

***Figure 3.** Corrosion Rate of Mild steel with and without the Gardenia aqualla extract.*

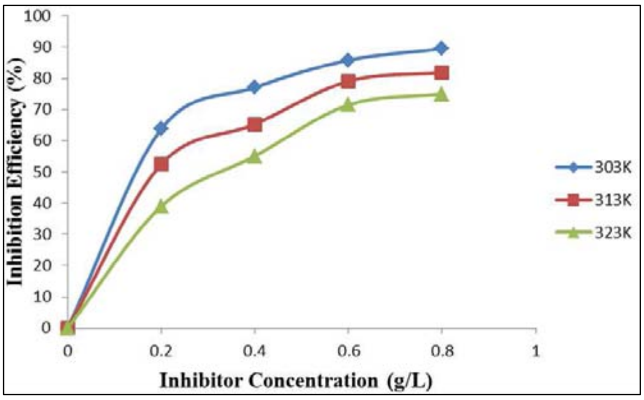

***Figure 4.** Inhibition Efficiency of Gardenia aqualla extract on Mild steel Corrosion in 1.0 M H_2SO_4.*

3.3. Adsorption Mechanism

The whole process of creating a barrier with the inhibitor or inhibiting the corrosion of mild steel can be said to occur through either the adsorption of *Gardenia aqualla* molecules or the formation of a layer of insoluble complex of the metal on the surface which acts as a barrier between the metal surface and the corrosive medium (this process is termed the mechanism of physisorption).

In addition, the neutral *Gardenia aqualla* may be adsorbed on the surface of the metal through the process of chemisorption involving the displacement of molecules of water from the metal surface and the sharing of electrons between iron and oxygen atoms.

As a result, Langmuir adsorption mechanism was used to control the inhibition reaction hence; it has the potential to physisorption occurrence as described below.

The concern of the mathematical relationship between surface coverage and concentration of inhibitor is expressed by Langmuir in the Equation below:

$$\frac{C}{\theta} = \frac{1}{k} + C$$

Where K is the equilibrium constant, C is the concentration of inhibitor. The plot in Figure 5 illustrates the Langmuir adsorption plot of $\frac{C}{\theta}$ versus C, which shows a straight line graph with the slope and R^2 values.

The Langmuir adsorption plot has linearity and its correlation coefficient is also good (the degree of fit between the experimental data and the isotherm equation) at different intervals of exposure. The values of R^2 are very close to unity, showing that the Langmuir adsorption isotherm was strongly obeyed [22], [23-24].

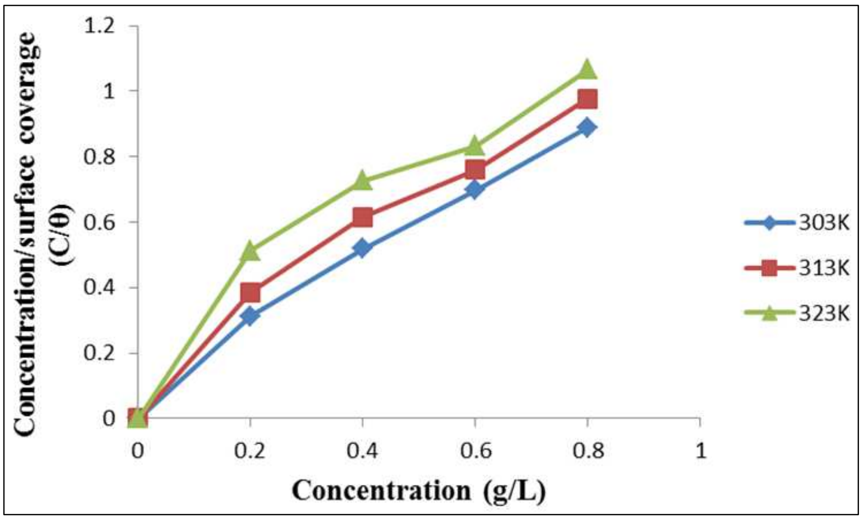

Figure 5. *Langmuir Isotherm Plot of the Corrosion of Mild steel in the presence of Gardenia aqualla extract.*

4. Conclusion

This study has illustrated that *Gardenia aqualla* extract prevented the corrosion of mild steel in 1 M H_2SO_4 solution at different temperatures. The efficiency of inhibition at different concentrations verified an optimum value of about 89.5% which increases on increasing the concentration of inhibitor and decreases with increase in temperature. The Langmuir isotherm was obeyed, which confirms the mechanism of physisorption as the process that aided corrosion inhibition of mild steel. *Gardenia aqualla* acted as a potential inhibitor for the corrosion of mild steel in 1 M H_2SO_4 solution.

Appendix

Table 2. *Summary of average weight loss (mg), corrosion rate (mm/yr), inhibition efficiency (%), surface coverage (θ) and concentration/surface coverage* $\frac{C}{\theta}$.

Temperature (K)	Concentration (g/100ml)	Weight loss (mg)	Inhibition efficiency (*I*) (%)	Corrosion rate (mm/yr)	Surface coverage (θ)	Concentration/surface coverage $\frac{C}{\theta}$
303	Blank	215.4		62.4		0
	0.2	77.9	63.8	22.6	0.64	0.313
	0.4	49.3	77.1	14.3	0.77	0.519
	0.6	30.5	85.8	8.80	0.86	0.698
	0.8	22.6	89.5	6.50	0.90	0.889
313	Blank	316.3		91.6		0
	0.2	150.4	52.5	43.5	0.52	0.385
	0.4	109.8	65.3	31.8	0.65	0.615
	0.6	66.2	79.1	19.2	0.79	0.759
	0.8	57.2	81.9	16.6	0.82	0.976
323	Blank	393.2		113.8		0
	0.2	239.8	39.0	69.4	0.39	0.513
	0.4	176.6	55.1	51.1	0.55	0.727
	0.6	112.0	71.5	32.4	0.72	0.833
	0.8	98.2	75.0	28.4	0.75	1.067

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