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Gmelina arborea Leaf Extract as an Eco – friendly Inhibitor for Mild Steel Corrosion in Acidic Medium

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Abstract: The inhibitory properties of *Gmelina arborea* leaf extract (GALE) on mild steel corrosion in 1 M H_2SO_4 solution was investigated by gravimetric and hydrogen evolution methods. The gravimetric data reveal that the inhibition efficiency of GALE increased with increase in extract concentration but decreased with a rise in temperature. The hydrogen evolution data indicate a marked reduction in the hydrogen evolution rate by the extract. The calculated thermodynamic parameters reveal the endothermic nature of the corrosion inhibition process. The surface morphology of the mild steel coupons by SEM analysis reveals a smoother mild steel surface in the presence of GALE compared to the blank. Based on a decrease in inhibition efficiency with a rise in temperature as well as a higher activation energy (E_a) in the presence of the extract relative to the blank, physical adsorption process has been proposed for the adsorption of GALE onto mild steel surface. The adsorption of GALE onto mild steel surface obeys the Langmuir adsorption isotherm.

1. Introduction

In the chemical industry, large quantities of mineral acids are used as pickling liquor for descaling of metals. The application of corrosion inhibitors to the pickling liquor helps in reducing the attack of the acid on the metal during the pickling process. Most of the efficient corrosion inhibitors in use are synthesized organic compounds having heteroatoms like N, S, and O (Ita and Abakedi, 2006; Hmamou *et al.*, 2013; Ghazoui *et al.*, 2014; Abakedi *et al.*, 2020; Merimi *et al.*, 2021; Salim *et al.*, 2022; Alharthi *et al.*, 2022; Tanwer and Shukla, 2023;). It has been reported that most of these compounds are hazardous to humans and the environment (Manhattan, 1994; Zarrok *et al.*, 2012; Abakedi and Moses, 2016), hence the need to replace them with safer corrosion inhibitors.

Research efforts have led to the discovery of natural products, especially plant extracts, as corrosion inhibitors. The major advantages of plant extracts include low toxicity, ease of extraction, low cost and environmentally – friendly characteristics. Some leaf extracts reported as efficient corrosion inhibitors for mild steel in acidic media include *Maesobatrya barteri* (Abakedi *et al.*, 2016), *Falcaria vulgaris* (Alimohammadi *et al.*, 2023), *Anthonotha macrophylla* (Iloamaeke *et al.*, 2024), *Commelina diffusa* (Abakedi *et al.*, 2024a), *Mukia maderaspatana* (Radha *et al.*, 2024), *Cannabis Sativa* (Haddou *et al.*, 2025), and Cinnamon (Senthooran *et al.*, 2025). The search for efficient corrosion inhibitors to mitigate the corrosion of mild steel in acidic medium is ongoing.

Gmelina arborea (English name: beechwood or gmelina) is a fast-growing deciduous tree belonging to the family Lamiaceae. *G. arborea* extracts have been documented to possess antioxidant (Sinha *et al.*, 2006), anti – diabetic (Kulkarni and Veeranjanevulu, 2013), anti-bacterial (Kenneth *et al.*, 2019), and anti – pyretic (Nayak *et al.*, 2015) activities. Preliminary phytochemical screening of *G. arborea* leaf extract indicates the presence of steroids, triterpenoids, saponins, protein, phenolic compounds, flavonoids and carbohydrates (Chothami and Patel, 2012). Previous studies (Abakedi *et al.*, 2018) revealed that *G. arborea* root extract is a good inhibitor of mild steel corrosion in acidic medium. Limited work (Ayuba *et al.*, 2020; Emekwisia *et al.*, 2024) has been done on *G. arborea* leaf extract as a corrosion inhibitor for mild steel in acidic medium. The aim of this work is to assess the inhibitory effect of *G. arborea* leaf extract on mild steel corrosion in 1 M H_2SO_4 solution.

2. Methodology

2.1 Test materials

The mild steel sample used in this work had the following chemical composition (w/w %): Mn (0.85), C (0.12), Si (0.09), S (0.06), P (0.05) and Fe (98.83). It was mechanically press - cut into 4 cm x 4 cm coupons. The coupons were polished to mirror finish using different grades of silicon carbide papers, degreased in absolute ethanol, dried in acetone and stored in a moisture –free desiccators before use in corrosion studies.

2.2 Preparation of Gmelina arborea leaf extract

Mature leaves of *Gmelina arborea* were collected from a farm in Ibesikpo, Nigeria and authenticated by a plant taxonomist in the Department of Botany and Ecological Studies, University of Uyo, Uyo, Nigeria. *G. arborea* leaves were washed with deionized water and shade – dried for seven days. They were then ground to powder. The dried ground samples of *G. arborea* leaves were macerated with 90% ethanol for three days, with intermittent stirring, at room temperature in a glass trough with cover. The mixture was then filtered. The filtrate was evaporated in a water bath at 40 °C to constant weight to obtain the crude extract in a beaker. Different extract concentrations (0.5 g/L, 1.0 g/L, 1.5 g/L, and 2.0 g/L) were prepared in 1 M H_2SO_4 solution for both the gravimetric and hydrogen evolution studies.

2.3 Gravimetric Method

Pre – washed and weighed mild steel coupons were suspended by glass hooks and completely immersed in separate 100 ml beakers containing 1 M H_2SO_4 solution (blank) and different concentrations of *G. arborea* leaf extract (0.5 g/L, 1.0 g/L, 1.5 g/L, and 2.0 g/L) in a thermostatic water bath maintained at 30 °C, 40 °C, 50 °C and 60 °C, respectively, for four (4) hours. After four hours, the coupons were retrieved from the corrodent, scrubbed with bristle brush under running water. They were then dipped into acetone and air – dried before reweighing. The weight losses of the coupons, in the absence and presence of the extract, were recorded.

The weight losses were used in calculating the corrosion rate, CR (mg cm^{-2} hr^{-1}), using the **Eqn. 1** (Abakedi *et al.*, 2024b):

$$\mathrm{CR\ (mg\ cm^{-2}hr^{-1})} = \frac{\mathrm{W}}{\mathrm{A.t}} \qquad \textbf{Eqn. 1}$$

where W is the weight loss of mild steel coupon (mg), A is the surface area (cm^2) and t is the immersion time (hours).

The inhibition efficiency, I_{wL} (%), of mild steel was evaluated using **Eqn. 2**:

$$I_{WL}(\%) = \left(1 - \frac{w_1}{w_0}\right) \times 100 \qquad \textbf{Eqn. 2}$$

where w_1 and w_0 are weight losses of mild steel coupons in 1 M H_2SO_4 solution with and without inhibitor, respectively.

2.4 Hydrogen Evolution Method

The hydrogen evolution measurements (via a gasometric assembly) were performed as described in literature (Abakedi and Sunday, 2017; Abakedi *et al*., 2024b). The corrodent was 100 ml of 1 M H_2SO_4 solution. About 8.0 g mild steel coupon was dropped into the 1 M H_2SO_4 solution (blank) and the reaction vessel quickly corked. The volume of H_2 gas evolved from the corrosion reaction was recorded every 60 seconds for 60 minutes. The experiment was repeated using 0.5 – 2.0 g/L *G. arborea* leaf extract (GALE) in 1 M H_2SO_4 solution. The inhibition efficiency I_{HE} (%) was calculated using **Eqn. 3**:

$$I_{HE} = \left(1 - \frac{V_1}{V_0}\right) \times 100 \qquad \textbf{Eqn. 3}$$

where V_0 and V_1 are the hydrogen evolution rates of mild steel coupons in 1 M H_2SO_4 solution without and with inhibitor, respectively.

2.5 Surface Morphology Study

Mild steel coupons (4 cm × 4 cm) were abraded to mirror finish with different grades of silicon carbide papers, cleaned and dried. The morphologies of the surfaces of the coupons were scanned with FEI Nova NanoSEM 230 analyzer after immersion in 100 ml 1 M H_2SO_4 for four hours without and with 2.0 g/L GALE.

3. Results and Discussion

3.1 Effect of Gmelina arborea leaf extract concentration on inhibition efficiency

Gmelina arborea leaf extract (GALE) concentration inhibited the corrosion of mild steel in 1 M H_2SO_4 solution as shown in **Figure 1**. It is observed that, at a constant temperature, an increase in GALE concentration leads to an increase in the inhibition efficiency. The highest inhibition efficiency of 64.75% was obtained at extract concentration of 2.0 g/L at 30 ºC. Also, an increase in inhibition efficiency with increase in extract concentration implies that the adsorption of the extract on mild steel. surface is concentration – dependent (Abakedi *et al*., 2024b).

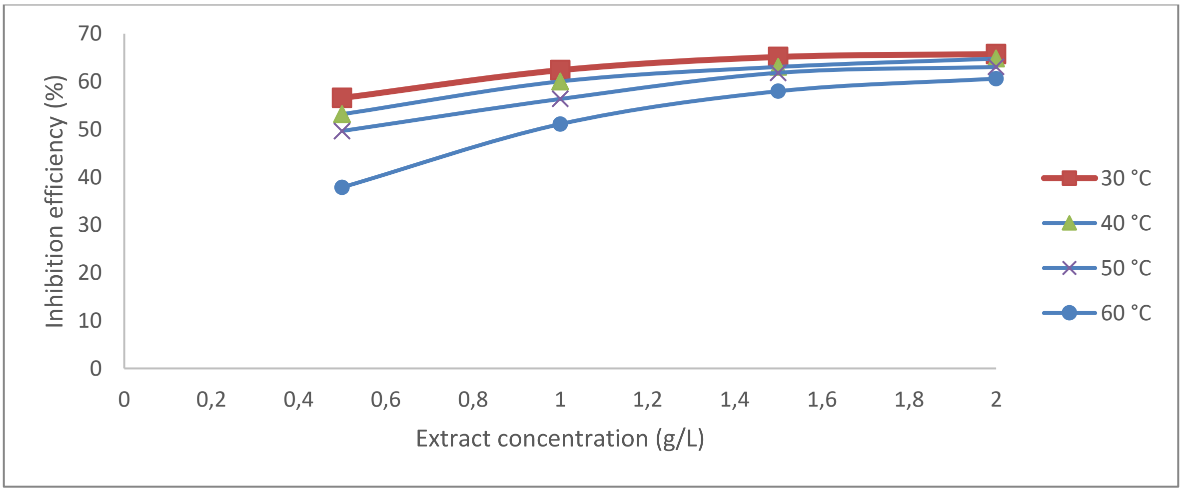


Figure 1. Effect of *G. arborea* leaf extract (GALE) concentration on the inhibition efficiency of mild steel in 1 M H_2SO_4 solution at 30 °C - 60 °C (Gravimetric measurements)

The results of the hydrogen evolution test are illustrated in **Figure 2**. It clearly reveals an appreciable reduction in the volume of hydrogen gas evolved in the presence of GALE relative to the blank. This indicates that the extract hindered the evolution of hydrogen gas. The inhibition efficiency by the hydrogen evolution method as presented in **Table 1** shows that the inhibition efficiency increases as GALE concentration increases. The highest inhibition efficiency by the hydrogen evolution method was 74.80% at 2.0 g/L extract concentration. The inhibition efficiencies by both gravimetric and hydrogen evolution methods followed a similar trend. The inhibitory effect of GALE could be attributed to the presence of phytochemicals in the extract. These phytochemicals adsorb onto the mild steel surface thereby reducing the attack by aggressive ions in solution.

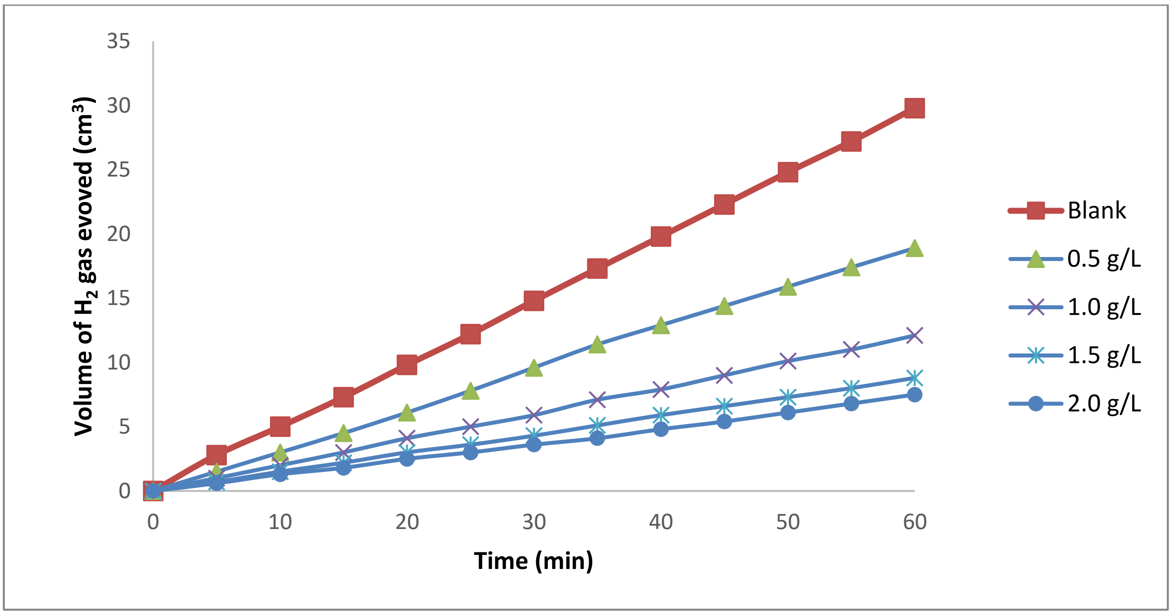


Figure 2. Variation of volume of H_2 gas evolved (cm^3) with time (min) for mild steel corrosion in 1 M H_2SO_4 in the absence and presence of *G. arborea* leaf extract at 30 °C

Table 1. Effect of *G. arborea* leaf extract concentration on inhibition efficiency of mild steel in 1 M H_2SO_4 solution at 30°C (Hydrogen evolution measurements)

Extract concentration (g/L)	H_2 evolution rate (cm^3/min)	Inhibition efficiency (%)
1 M H_2SO_4 (Blank)	0.497	-
0.5	0.315	36.62
1.0	0.212	57.84
1.5	0.147	70.42
2.0	0.125	74.85

3.2 ***Effect of Temperature on Inhibition Efficiency***

Temperature influenced the effectiveness of GALE as inhibitor for mild steel corrosion in acidic medium as presented in **Table 2**. It is observed that the inhibition efficiency of GALE decreases as the temperature rises. A decrease in inhibition efficiency with a rise in temperature implies a thermal desorption of the extract on the steel surface. Additionally, it indicates that GALE functions more effectively as an inhibitor at a lower temperature than at a higher temperature.

Table 2. Calculated values of corrosion rate and inhibition efficiency for mild steel in 1 M H_2SO_4 in the absence and presence of *G. arborea* leaf extract at 30 °C – 60 °C

Extract conc. (g/L)	Corrosion rate ($mg\ cm^{-2}\ hr^{-1}$)				Inhibition efficiency (%)			
	30 °C	40 °C	50 °C	60 °C	30 °C	40 °C	50 °C	60 °C
Blank	2.5557	4.9063	7.0000	15.6719	-	-	-	-
0.5	1.1094	2.2969	3.5234	9.7344	56.57	53.18	47.67	37.89
1.0	0.9609	1.9609	3.0547	7.6641	62.39	60.03	56.36	51.10
1.5	0.8906	1.8125	2.6719	6.5859	65.14	63.06	61.83	57.98
2.0	0.8750	1.7266	2.5859	6.1719	65.75	64.81	63.06	60.62

The activation energies (E_a) of the corrosion process in the absence and presence of GALE, respectively, were calculated using the Arrhenius equation (**Eqn. 4**):

$$\ln CR = \frac{-E_a}{RT} + \ln A \qquad \textbf{Eqn. 4}$$

where CR is the corrosion rate, R is the universal gas constant, T is the temperature in Kelvin while A is the pre-exponential factor.

From the gradients of ln CR vs. 1/T plots (**Figure 4**), the activation energies (E_a) of mild steel corrosion in 1 M H_2SO_4, in the absence (blank) and presence of GALE, were evaluated and the results presented in **Table 3**. **Table 3** reveals that the E_a values in the presence of GALE are higher than the E_a of the blank (48.20 kJ mol^{-1}). A higher E_a value in the presence of the extract relative to the blank implies that the extract increased the energy barrier of the corrosion reaction and hence slowed down the corrosion process. Furthermore, a higher E_a value in presence of the extract relative to the blank signifies a physisorption process (Ita *et al.*, 2013; Ashmawy *et al.*, 2023).

The values of enthalpy of activation ($\Delta H°_{ads}$) and entropy of activation ($\Delta S°_{ads}$) were calculated from the Transition State equation (**Eqn. 5**) (Uzah *et al.*, 2023):

$$\left(\frac{CR}{T}\right) = \left(\frac{R}{Nh}\right) \exp\left(\frac{\Delta S^{\circ}_{ads}}{R}\right) \exp\left(\frac{-\Delta H^{\circ}_{ads}}{RT}\right) \qquad \textbf{Eqn. 5}$$

where CR is the corrosion rate, T is temperature in Kelvin, R is the universal gas constant, N is Avogadro's number while h is the Planck's constant.

Plots of ln (CR/T) vs. 1/T give straight lines (Figure 5). The values of $\Delta H°_{ads}$ and $\Delta S°_{ads}$ presented in Table 3 were obtained from the gradients ($-\Delta H°_{ads}/R$) and intercepts of [ln (R/Nh) + $\Delta S°_{ads}/R$] of the plots. The positive values of $\Delta H°_{ads}$ both in the absence and presence of GALE is indicative of the endothermic nature of corrosion process. Additionally, the negative values of $\Delta S°_{ads}$ obtained signify a decrease in the disorderliness of the system.

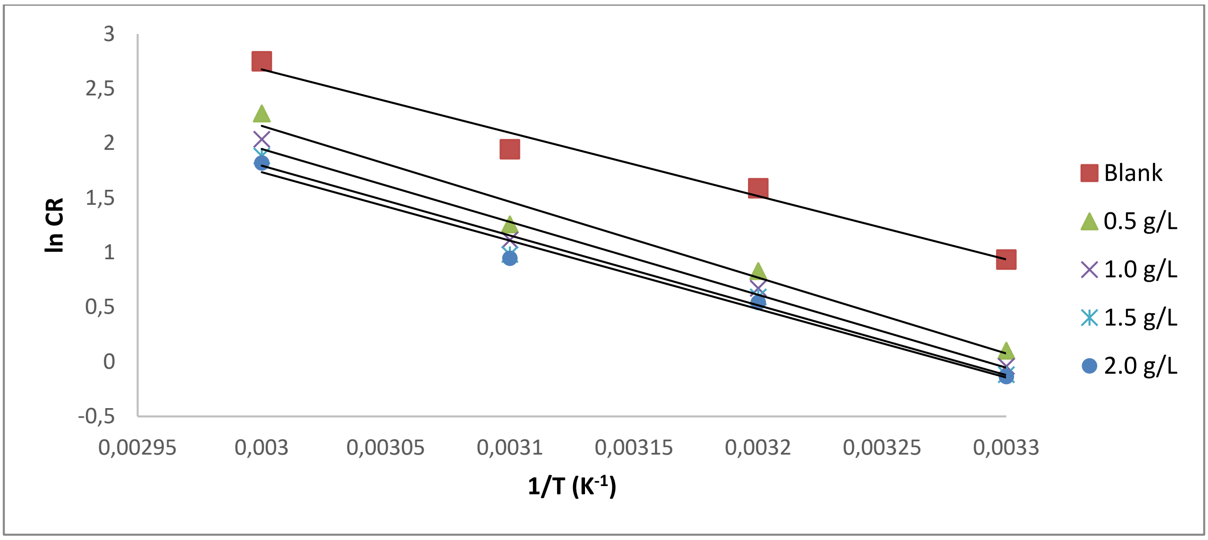


Figure 3: Arrhenius plot for mild steel corrosion in 1 M H_2SO_4 in the absence and presence of GALE

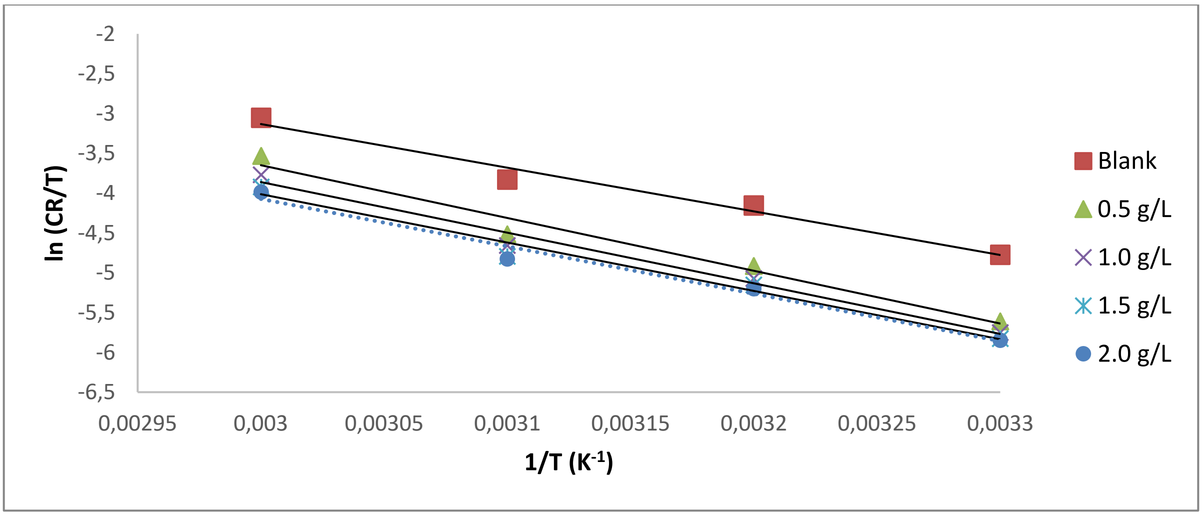


Figure 4: Transition State plot for mild steel corrosion in 1M H_2SO_4 in the absence and presence of GALE

Table 3: Calculated values of thermodynamic parameters for mild steel corrosion in 1 M H_2SO_4 in the absence and presence of *G. arborea* leaf extract

Extract conc. (g/L)	E_a (kJ mol^{-1})	ΔH (kJ mol^{-1})	ΔS (JK^{-1}mol^{-1})
1 M H_2SO_4 **(**Blank)	48.20	45.58	-86.83
0.5	57.73	55.11	-62.55
1.0	55.48	52.86	-71.06
1.5	52.13	50.51	-79.36
2.0	52.08	49.47	-82.99

3.3 Adsorption Studies

The best fit for the adsorption of GALE onto mild steel surface was obtained with the modified Langmuir adsorption isotherm defined by (**Eqn. 6**) (Villani *et al.*, 1999; Ran *et al.*, 2023):

$$\frac{C}{\theta} = \frac{n}{K_{ads}} + nC \qquad \text{Eqn 6}$$

where C is the concentration of inhibitor, θ is the degree of surface coverage, and K_{ads} is the equilibrium constant of the adsorption process.

Fig. 5 reveals linear plots of C/θ vs. C indicating that the adsorption of GALE onto mild steel surface obeyed the Langmuir adsorption isotherm. The values of K_{ads}, evaluated from the intercepts of the graph, are presented in Table 4. It is observed that the highest value of K_{ads} was obtained at 30 °C while the lowest occurred at 60 °C. Additionally, the values of K_{ads} decrease with a rise in temperature from 30 °C to 60 °C. A decrease in the value of K_{ads} as temperature rises signifies that the extract was most strongly adsorbed on mild steel surface at 30 °C. A decrease in the value of K_{ads} as well as a decrease in the inhibition efficiency with a rise in temperature of the system supports a physical adsorption (physisorption) process.

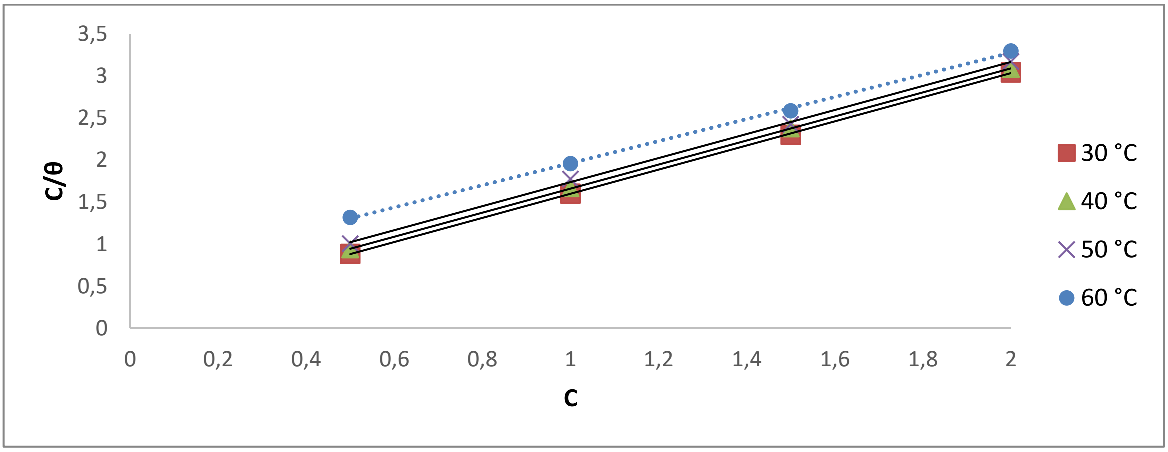


Figure 5: Langmuir isotherm for mild steel corrosion in 1 M H_2SO_4 in the absence and presence of GALE at 30 °C- 60 °C

Table 4: Some parameters of the linear regression of Langmuir adsorption isotherm for mild steel corrosion in 1 M H_2SO_4 containing GALE

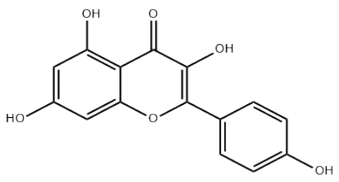
Temperature	R^2	n	K_{ads} (L g^{-1})
303 K	0.9999	1.435	8.729
313 K	1.0000	1.430	6.212
323 K	0.9991	1.429	4.640
333 K	0.9992	1.314	2.026

Literature shows that chemical composition of Gmelina arborea leaf extract is rich in various components as alkaloids, Tanins, Flavonoids, etc. as confirmed by Lawson *et al*., 2016 (**Table 5**). Ghareeb *et al*., isolated seven flavonoidal compounds: ; luteolin (1), luteolin-4'-O-D-C-galactoside (2), kaempferol (3), quercetin-4, 1,3-O-D-C-glucopyranoside (isoquercitrin) (4), quercetin-3-O-C-L-rhamnopyranosyl-(1'"6")-4,1, 1,4-O-D-C-glucopyranoside (rutin) (5), luteolin-7-O-D-C-galactoside (6) and quercetin-3-O-C-L- 4,4,1,1,14 rhamnopyranosyl-(1''' 6")-C-D-galactopyranoside (quercetin-3-O-robinobioside) (7). Their structures were elucidated via UV, IR and NMR spectral techniques as well as (Co-PC, Co-TLC and Co-m.p.) and acid hydrolysis. Their antioxidant activity (AOA) was evaluated via 1,1'-diphenyl-2-picraylhydrazyl free radical (DPPH) and phosphomolybdenum assays, while their cytotoxic activity was evaluated toward liver-carcinoma cell line (HepG-2) via Sulphorhodamine-B assay (Ghareeb *et al*., 2014).

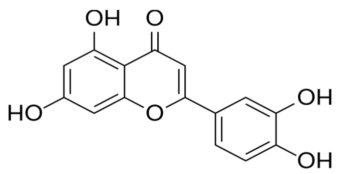
Table 5. Phytochemical analysis of G. arborea leaves (Lawson *et al*., 2016).

Chemical families	Expression
Alkaloids	+
Tannins: 1. Catechin; 2. Gallic	++; +/-
Flavonoids	-
Anthocyanins	+/-
Leucoanthocyanins	+/-
Mucilage	++
Reducing sugars	-
Cyanogenic glycosides	-
Quinones	-
Saponins	++
Sterols and triterpenes	-
Anthracen glycosides	-
Coumarins	-

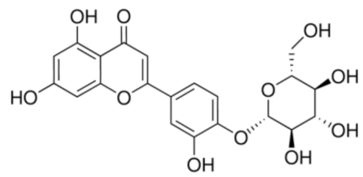
(+) detected, (++) abundantly detected, (+/-) weakly detected, (-) absent.



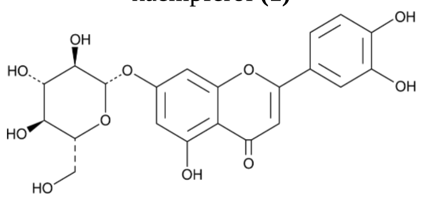
kaempferol **(1)**



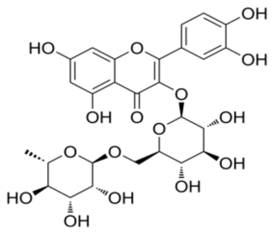
luteolin **(2)**



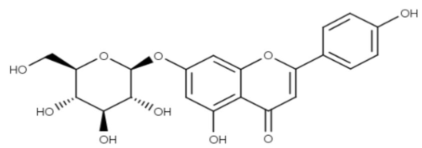
luteolin-4'-O-βD4C1-galactopyranoside **(3)**



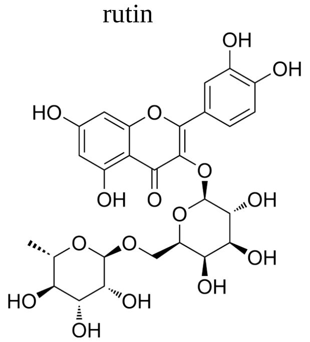
leteolin-7-galactoside **(4)**



rutin



luteolin-7-O- -D- C -galactoside

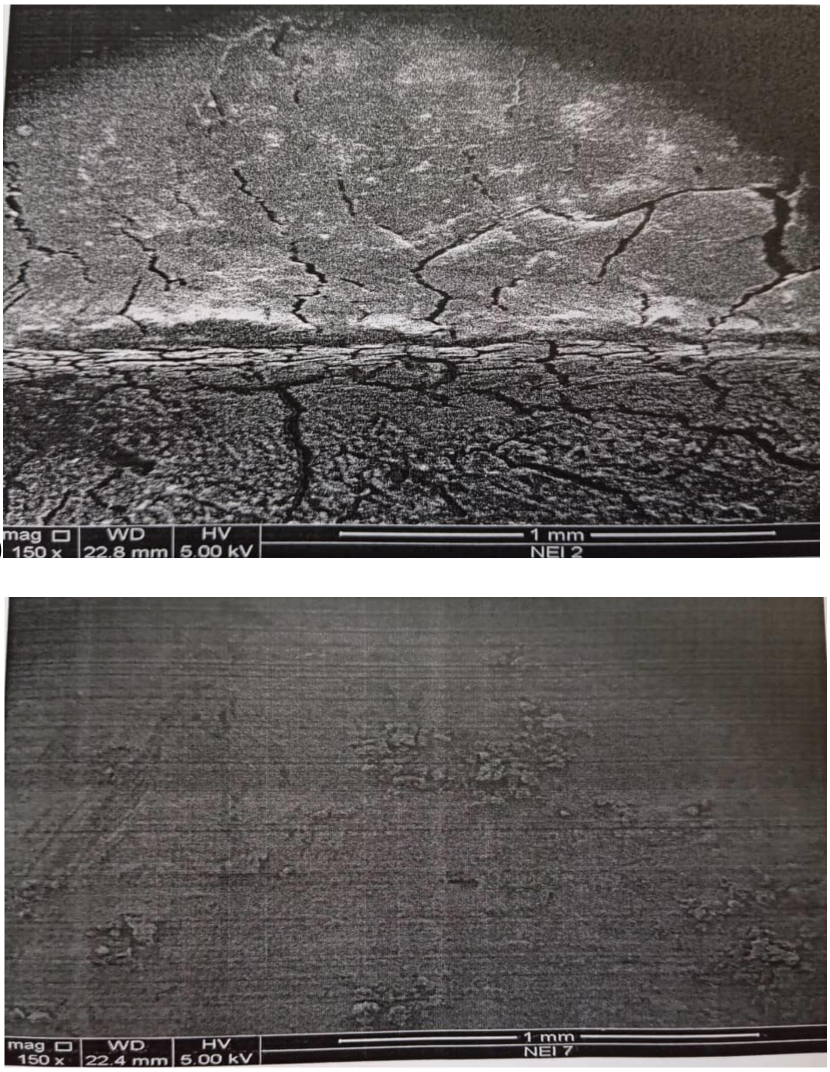


quercetin-3-O-robinobioside)

These various and numerous molecules are rich in aromatic cycles, heteroatoms... and can easily adsorb on the surface of the metal and create a barrier against the arrival of H^+ ions and consequently, block the degradation process of the metal. This competitive or competing contribution allows the introduction of the notion of the effect of intermolecular synergy discussed by several authors (Ali *et al*., 2014; Rostami *et al*., 2018; Salim *et al*., 2022; Lrhoul *et al*., 2023; Sanjay *et al*., 2024).

3.4 SEM Analysis

The surface morphology of mild steel coupons immersed in 1 M H_2SO_4 in the absence and presence of 2 g/L GALE, as analyzed by Scanning Electron Microscopy (SEM), are presented in **Figure. 6** (a and b). Figure 6(a) indicates the presence of cracks on mild steel surface corroded in 1 M H_2SO_4 solution (blank) due to severe corrosion while Figure 6(b) reveals a smoother mild steel surface in the presence of 2.0 g/L GALE is smooth. This signifies that the extract protected the mild steel surface from attack by aggressive ions in solution by adsorbing onto it.



(a)

(b)

Figure 6: SEM micrographs of mild steel in 1 M H_2SO_4 in the (a) absence of extract (b) presence of GALE

Conclusion

On the basis of this work, the following conclusions could be drawn:

i *Gmelina arborea* leaf extract (GALE) appreciably inhibited the corrosion of mild steel corrosion in sulphuric acid solution. It could be incorporated as an eco–friendly inhibitor into a pickling liquor.

ii The inhibition efficiency increases as the concentration of GALE increases but decreases as temperature rises, signifying a stronger adsorption between the mild steel surface and the extract at lower temperature than at higher temperature.

iii The E_a values in the presence of GALE being greater than the value in the blank coupled with a decrease in inhibition efficiency as temperature rises, is indicative of a physical adsorption mechanism.

iv The calculated values of ΔH°_{ads} for mild steel corrosion containing GALE are positive, revealing the endothermic nature of the corrosion inhibition process.

v. The adsorption of GALE onto mild steel surface obeyed the Langmuir adsorption isotherm, though the K_{ads} values indicate that adsorption bond between GALE and mild steel surface weakens as temperature rises.

vi. SEM analyses indicate that GALE offered a better protection for mild steel surface in acidic medium than the blank, probably by shielding the mild steel surface from attack by aggressive ions.

COMPETING INTEREST: The authors declare that there are no conflicts of interest

Disclosure statement: *Conflict of Interest:* The authors declare that there are no conflicts of interest.

Compliance with Ethical Standards: This article does not contain any studies involving human or animal subjects.

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