

Inhibition of Mild Steel Corrosion by *Gmelina arborea* Root Extract in 1 M H₂SO₄ Solution

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Original Research Article

ABSTRACT

The inhibition of mild steel corrosion in sulphuric acid solution by *Gmelina arborea* root extract was studied using weight loss and hydrogen evolution methods. The results obtained reveal that the extract appreciably inhibited the corrosion of mild steel in the acid medium. The inhibition efficiency increased with increase in *Gmelina arborea* root extract concentration but decreased with increase in temperature. The highest inhibition efficiency of 78.59% occurred at extract concentration of 2 g/L at 30°C by weight loss measurements. Physical adsorption has been proposed for the adsorption of *Gmelina arborea* root extract onto the mild steel surface. The calculated thermodynamic parameters revealed that the adsorption process was endothermic and spontaneous. The phytochemical screening of *Gmelina arborea* root extract revealed the presence of alkaloids, saponins, anthraquinones and terpenes. The adsorption of the root extract on mild steel surface obeyed the Langmuir adsorption isotherm.

Keywords: *Gmelina arborea*; corrosion inhibition; physisorption; extract; Langmuir isotherm.

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1. INTRODUCTION

Pure metals and its alloys have gained extensive use in industrial and engineering applications. These metals have been deployed into various service environments which are either acidic or alkaline. Upon exposure to these aggressive environments, the metals experience weakening in their mechanical strength or complete structural failure due to corrosion [1]. Annual economic losses that result from corrosion processes is of global concern. Due to an increasing need for the use of metals for engineering infrastructure, corrosion inhibitors are desired in order to prolong the service life of metallic components in contact with aggressive environments. Numerous organic compounds have been synthesised to tackle this problem [2-6]. These organic compounds contain nitrogen, oxygen, sulphur and/or phosphorus. Researchers nowadays do extensive work to curb the situation by using green inhibitors because the synthetic organic compounds are toxic, non-biodegradable and environmentally unfriendly [7-10]. Reports have documented the use of extracts from *Diospyros mespiliformis* [11], *Cucurbita maxima* [12], *Piper longum* [13], *Chromolaena odorata* [14], African breadfruit [15] and *Hibiscus sabdariffa* [16] as potential green inhibitors of mild steel corrosion in acidic medium.

Gmelina arborea (Family: Verbenaceae) is a tropical, evergreen perennial deciduous tree that grows to a height of 12 to 30 m. It grows preferably in the moist fertile area. It is moderately adaptable and survives well on a wide range of soil types: acid soils, calcareous loams, and lateritic soils [17]. In Nigeria, the stem bark is boiled as a concoction for the treatment of chest and waist pain, lumbago and rheumatism. Extracts of *Gmelina arborea* are used in herbal medicine for its analgesic [18] anti-diabetic [19], antimicrobial [20], antioxidant [21] and wound healing properties [22]. Previous studies [23] revealed that *Gmelina arborea* stem bark extract appreciably inhibits mild steel corrosion in acidic medium. The aim of this work was to study the inhibitory effect of *Gmelina arborea* root extract on mild steel corrosion in H₂SO₄ solution.

2. MATERIALS AND METHODS

2.1 Test Materials

The mild steel sheet was purchased from Kenjohnsons Limited, Uyo, Akwa Ibom State, Nigeria. It had the following chemical

composition (wt. %): C (0.12), Mn (0.85), S (0.06), P (0.05), Si (0.09) and Fe (98.83). The sheet was mechanically pressed - cut into 4.0 cm x 4.0 cm (for weight loss method) and 2.0 cm x 4.0 cm (for hydrogen evolution studies) coupons, respectively. The coupons were abraded with silicon carbide papers to mirror finish, washed in absolute ethanol and acetone, dried in room temperature and stored in moisture-free desiccators before use in corrosion studies. All solutions were prepared using deionized water.

2.2 Preparation of *Gmelina arborea* Root Extract

Fresh roots of *Gmelina arborea* were collected from a farm in Urua Ekpa, Akwa Ibom State, Nigeria. They were washed, cut into small pieces and air - dried for seven days. The roots were ground to powder form. The *Gmelina arborea* root extract was obtained following the standard procedure reported previously [24]. The root extract was used for the preparation of 0.5 g/L, 1.0 g/L, 1.5 g/L and 2.0 g/L extract concentrations, respectively, in 1 M H₂SO₄ solution.

2.3 Phytochemical screening of *Gmelina arborea* Root Extract

The qualitative phytochemical analysis of *Gmelina arborea* root extract was done using standard procedures [25 - 26].

2.4 Weight Loss Method

Weighed mild steel coupons were immersed in 100 ml of 1 M H₂SO₄ solution (blank) in open beakers with the aid of glass rods and hooks. One mild steel coupon per beaker was used. The temperature of the experiments was maintained at 30°C, 40°C, 50°C, and 60°C, respectively, by placing the beakers in a thermostatic water bath. The mild steel coupons were retrieved after 4 hours intervals and washed in running water, rinsed in acetone and allowed to dry at room temperature before reweighing. The experiments were also repeated in 1 M H₂SO₄ solution containing 0.5 g/L, 1.0 g/L, 1.5 g/L, and 2.0 g/L *Gmelina arborea* root extract concentrations, respectively.

The corrosion rate (CR) was calculated using the formula [9]:

$$CR \text{ (mg cm}^{-2}\text{hr}^{-1}\text{)} = \left(\frac{W}{At} \right) \quad (1)$$

where W is the weight loss (mg), A is the total surface area (cm^2) while t is the exposure time (hours).

The inhibition efficiency $I(\%)$ was evaluated through equation (2) [1]:

$$I(\%) = \left(\frac{W_0 - W_1}{W_0} \right) \times 100 \quad (2)$$

where W_0 and W_1 are the weight losses of the mild steel coupons in the absence and presence of extract, respectively, in 1 M H_2SO_4 at the same temperature.

2.5 Hydrogen Evolution Method

The hydrogen evolution method (via a gasometric assembly) is a rapid and sensitive technique used to monitor the rate of corrosion through the rate of hydrogen gas evolved from a metal corroding in aqueous media [27-28]. The reaction vessel and procedure for the corrosion process by this method are as described by other workers [7]. A 100 ml of 1 M H_2SO_4 solution was transferred into the reaction vessel. Mild steel coupons weighing 6 g were dropped into the 1 M H_2SO_4 solution (blank). The volume of H_2 gas evolved from the corrosion reaction was recorded every 60 seconds for 60 minutes at 30°C . The experiment was repeated in the presence of 0.5 g/L – 2.0 g/L *Gmelina arborea* root extract (inhibitor) in 1 M H_2SO_4 solution.

The inhibition efficiency $I(\%)$ by the gasometric method was calculated using Equation (3) [29]:

$$I(\%) = \left(1 - \frac{R_{H1}}{R_{H0}} \right) \times 100 \quad (3)$$

where R_{H0} and R_{H1} are the H_2 gas evolution rates in the absence and presence of extract, respectively, at a specific time.

2.6 Scanning Electron Microscopy

The polished and cleaned mild steel coupons were immersed for four (4) hours in 1 M H_2SO_4 solution (blank) and in 1 M H_2SO_4 solution containing 2.0 g/L *Gmelina arborea* root extract, respectively, at 30°C . The coupons were cleaned as described in Section 2.4. A scanning electron microscope (SEM) was used to analyse the morphology of the mild steel surfaces.

3. RESULTS AND DISCUSSION

3.1 Results of Phytochemical Analysis of *Gmelina Arborea* Root Extract

The results of the qualitative phytochemical analysis of *Gmelina arborea* ethanol root extract indicate the presence of alkaloids, saponins, anthraquinones and terpenes.

3.2 Effect of *Gmelina arborea* Root Extract Concentration on Inhibition Efficiency

The inhibition efficiency of *Gmelina arborea* root extract on mild steel corrosion in 1 M H_2SO_4 solution increases with increase in the extract concentration (Fig. 1). The highest inhibition efficiency of 78.59% was obtained at an extract concentration of 2.0 g/L at 30°C by the weight loss method (Table 1). Fig. 2 depicts a drastic reduction in the volume of H_2 gas evolved in mild steel corrosion in 1 M H_2SO_4 solution in the presence of *Gmelina arborea* root extract compared to the blank. The reduction in the volume of H_2 gas evolved increased with increase in the root extract concentration, indicating that the extract inhibited the corrosion of mild steel in the medium. The calculated values of inhibition efficiency by the hydrogen evolution measurements (Table 2) reveal that the inhibition efficiency increased with increase in the extract concentration. A similar trend was observed in the inhibition efficiencies obtained by both the weight loss and hydrogen evolution measurements. This reveals that *Gmelina arborea* root extract is a good inhibitor of mild steel corrosion in sulphuric acid solution.

3.3 Effect of Temperature on Inhibition Efficiency

Table 2 reveals that the inhibition efficiency decreases with increase in temperature, suggesting that an increase in temperature increases the degree of inhibitor desorption from the surface of the mild steel, especially at high temperatures.

The activation energy (E_a) for mild steel corrosion in 1 M H_2SO_4 solution in the absence and presence of *Gmelina arborea* root extract was evaluated using the Arrhenius equation [24]:

$$\ln CR = \frac{-E_a}{RT} + \ln A \quad (4)$$

where CR is the corrosion rate, R is the universal

gas constant, T is the absolute temperature, E_a is factor.
activation energy while A is the pre-exponential

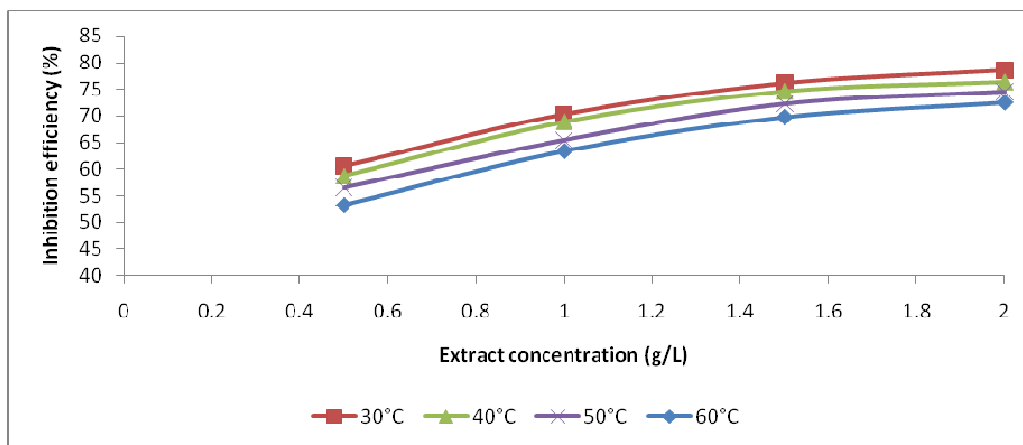


Fig. 1. Effect of *Gmelina arborea* root extract concentration (g/L) on the inhibition efficiency (%) of mild steel corrosion in 1 M H_2SO_4 solution at different temperatures

Table 1. Calculated values of corrosion rate and inhibition efficiency for mild steel corrosion in 1 M H_2SO_4 in the absence and presence of different concentrations of *Gmelina arborea* root extract

Extract concentration	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	2.554	4.906	7.000	16.141	-	-	-	-
0.5 g/L	1.007	2.023	3.039	7.547	60.55	58.76	56.58	53.24
1.0 g/L	0.757	1.523	2.414	5.906	70.34	68.95	65.51	63.41
1.5 g/L	0.609	1.242	1.937	4.875	76.15	74.68	72.32	69.80
2.0 g/L	0.547	1.156	1.781	4.429	78.59	76.43	74.55	72.56

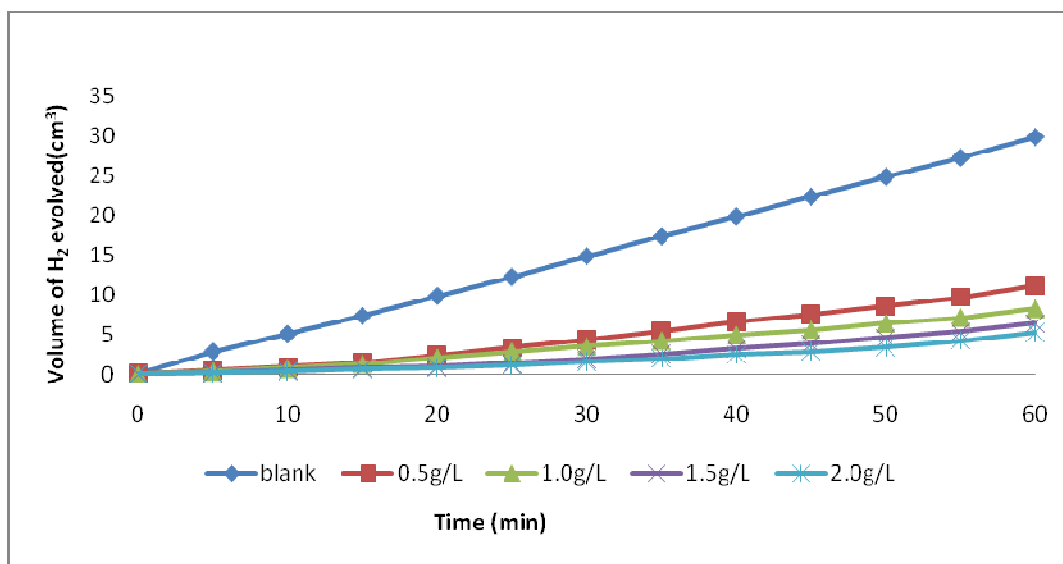


Fig. 2. Variation of the volume of H_2 gas evolved (cm^3) with time (min) for mild steel corrosion in 1 M H_2SO_4 in the absence and presence of *Gmelina arborea* root extract at 30°C

Table 2. Effect of *Gmelina arborea* root extract concentration on inhibition efficiency of mild steel in 1 M H₂SO₄ solution at 30°C (Hydrogen evolution measurements)

Extract concentration (g/L)	H ₂ evolution rate (cm ³ min ⁻¹)	Inhibition efficiency (%)
Blank	0.4967	-
0.5	0.1867	62.42
1.0	0.1383	72.15
1.5	0.1067	78.52
2.0	0.0867	82.55

Fig 3 illustrates a linear plot of $\ln CR$ vs $1/T$ for the mild steel corrosion in 1 M H₂SO₄ solution in the absence and presence of the extract. The E_a values presented in Table 3 were evaluated from the gradients of the plot. It is observed that the E_a values in H₂SO₄ – *Gmelina arborea* root extract medium were higher than that of the blank (47.36 kJ mol⁻¹). When there is an increase in activation energy (E_a) in the presence of extract compared to the blank, it is an indication of physical adsorption (physisorption) and when there is a decrease in the E_a value in the presence of the extract compared to the blank, chemical adsorption is implied [30 - 31]. In this study, where the E_a values in the presence of the

extract are higher than that of the blank. The adsorption of *Gmelina arborea* root extract on mild steel surface is therefore proposed to occur via physisorption mechanism. This assertion is further supported by a decrease in inhibition efficiency as temperature increases.

The values of entropy of activation (ΔS°_{ads}) and enthalpy of activation (ΔH°_{ads}) also presented in Table 3 were obtained from the alternative formulation of the transition state equation [10]:

$$\ln\left(\frac{CR}{T}\right) = \left[\ln\left(\frac{R}{Nh}\right) + \frac{\Delta S^\circ_{ads}}{R}\right] - \frac{\Delta H^\circ_{ads}}{RT} \quad (5)$$

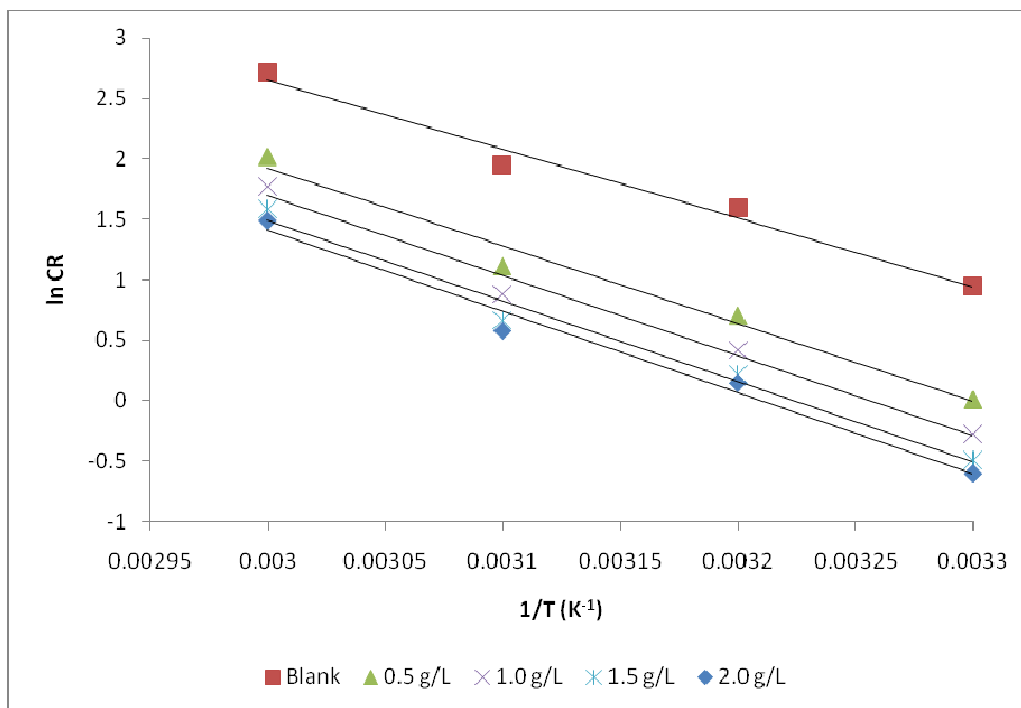


Fig. 3. Arrhenius plot for mild steel corrosion in 1 M H₂SO₄ solution in the presence and absence of *Gmelina arborea* root extract, respectively

Table 3. Calculated values of thermodynamic parameters for mild steel corrosion in 1 M H₂SO₄ in the absence and presence of *Gmelina arborea* root extract

Extract concentration	E_a (kJ mol ⁻¹)	ΔH_{ads}° (kJ mol ⁻¹)	ΔS_{ads}° (JK ⁻¹ mol ⁻¹)
1 M H ₂ SO ₄	47.36	46.29	-84.54
0.5 g/L	53.60	50.96	-76.92
1.0 g/L	55.04	52.40	-74.56
1.5 g/L	55.59	52.92	-74.65
2.0 g/L	55.77	53.13	-74.74

where T is the absolute temperature, CR is the corrosion rate, h is the Planck's constant, and N is the Avogadro's number and R is the universal gas constant. The plots of $\ln(CR/T)$ against $(1/T)$ is shown in Fig 4. This plot has gradient of $(-\Delta H_{ads}^{\circ}/R)$ and intercepts of $[\ln(R/Nh) + (\Delta S_{ads}^{\circ}/R)]$ where the values of ΔS_{ads}° and ΔH_{ads}° were obtained. Positive values of ΔH_{ads}° reveal that the adsorption of *Gmelina arborea* root extract onto mild steel surface was an endothermic process.

3.4 Adsorption Studies

The Langmuir adsorption isotherm proposes that adsorption occurs on specific homogenous sites on the metal surface and is used successfully in many monolayer adsorption processes [7]. The modified Langmuir isotherm was used to investigate the mechanism of adsorption, which is expressed in Equation (6) [32]:

$$\frac{C}{\theta} = \frac{n}{K_{ads}} + nC \quad (6)$$

where θ is the degree of surface coverage, C is the inhibition concentration, and K_{ads} is the equilibrium adsorption constant. The plot of C/θ against C (Fig 5) is a straight line graph which shows that the adsorption of *Gmelina arborea* root extract on mild steel in 1 M H₂SO₄ obeys the Langmuir adsorption isotherm. The values of K_{ads} which indicate the adsorption strength of the inhibitor onto the metal surface [7], was evaluated by the intercept of the graph. The standard free energy of adsorption ΔG_{ads}° is determined from the equation (7) [29]:

$$\Delta G_{ads}^{\circ} = -RT \ln(55.5K_{ads}) \quad (7)$$

where K_{ads} is the equilibrium adsorption constant, ΔG_{ads}° is the standard free energy of adsorption and the value 55.5 is the molar concentration of water in solution expressed in mol/dm³.

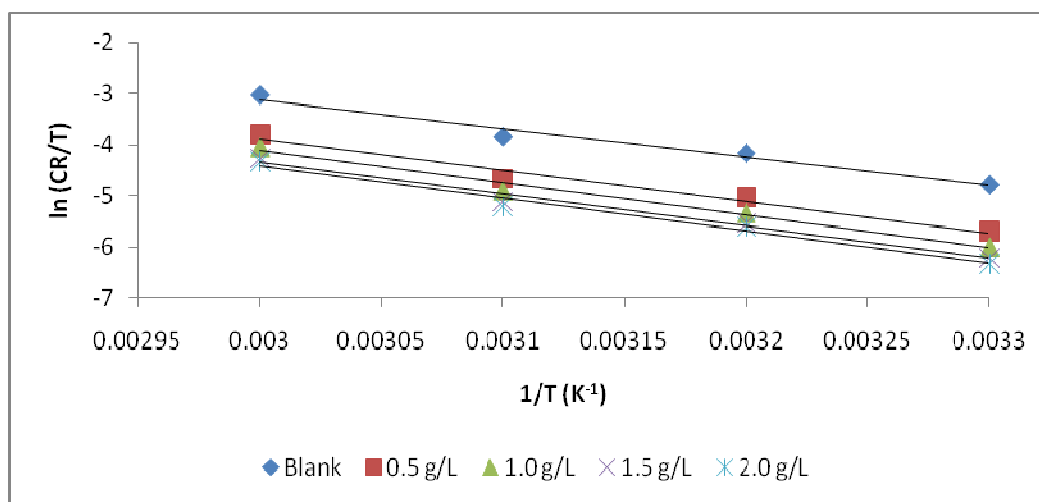


Fig. 4. Transition state plot for mild steel corrosion in 1 M H₂SO₄ solution in the presence and absence of *Gmelina arborea* root extract

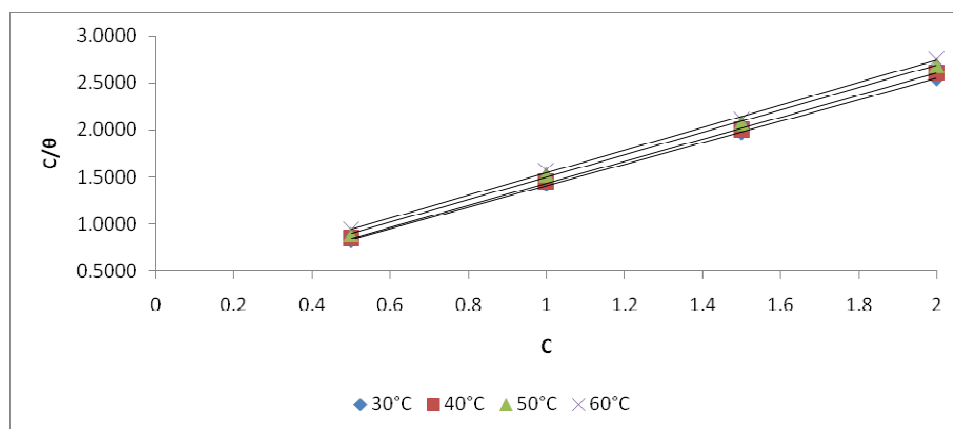


Fig. 5. Langmuir isotherm plot for mild steel corrosion in 1 M H₂SO₄ solution containing *Gmelina arborea* root extract at 30°C – 60°C

The values of K_{ads} and ΔG°_{ads} are represented in Table 4. Values of ΔG°_{ads} less negative than -20 kJ mol^{-1} reveal electrostatic interaction between charged organic molecules and the charged metal surface (physisorption) while values of ΔG°_{ads} more negative than -40 kJ mol^{-1} reveal charge sharing or transfer from the organic molecules to the metal surface to form a coordinate bond (chemisorption) [33]. The negative values of ΔG°_{ads} indicate the spontaneity of adsorption of *Gmelina arborea* root extract onto the mild steel surface.

3.5 Surface Morphology Analysis

Scanning electron microscope (SEM) images were taken in order to study the surface

morphology of mild steel in the absence and presence of *Gmelina arborea* root extract, respectively. SEM micrograph of the surface of mild steel after 4 hours of immersion in 1 M H₂SO₄ solution in the absence of *Gmelina arborea* extract (Fig. 6a) reveals that the mild steel surface was severely corroded. Cracks could be seen on the mild steel surface. On the contrary, SEM micrograph of mild steel in the presence of 2.0 g/L *Gmelina arborea* root extract (Fig. 6b) reveals a smoother surface. This may be attributed to the formation of an adsorbed film of *Gmelina arborea* molecules on a mild steel surface, which inhibit the corrosion of mild steel in 1 M H₂SO₄ solution.

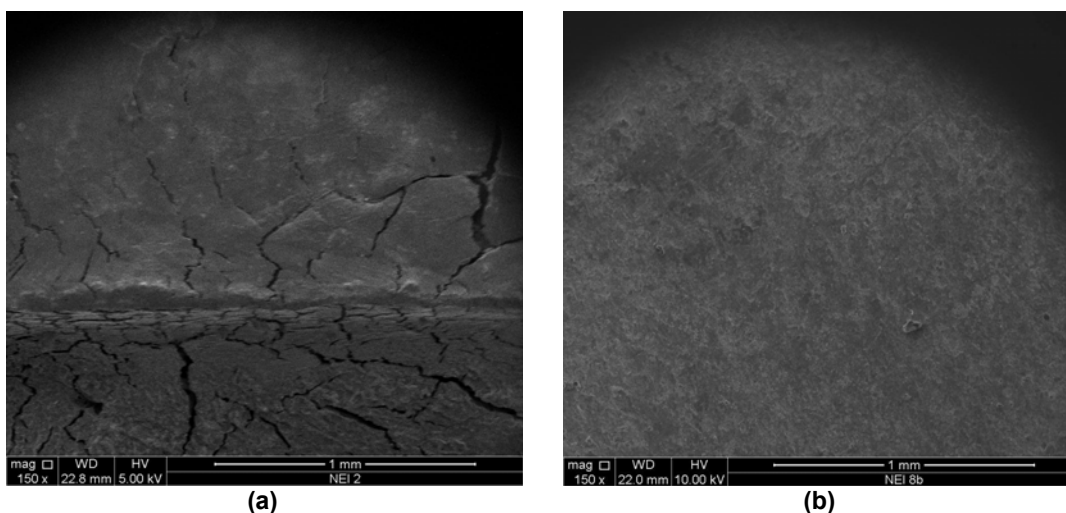


Fig. 6. SEM micrograph of mild steel specimens after 4 hours of immersion in (a) 1 M H₂SO₄ solution (blank) and (b) 1 M H₂SO₄ solution containing 2.0 g/L *Gmelina arborea* root extract at 30°C

Table 4. Some parameters of the linear regression of Langmuir adsorption isotherm for mild steel corrosion in 1 M H₂SO₄ solution containing *Gmelina arborea* root extract

Temperature	R ²	n	1/K _{ads} (g/L)	K _{ads} (L/g)	ΔG ^o _{ads} (kJ mol ⁻¹)
303K	0.9998	1.14	0.2643	3.7835	-13.469
313K	0.9997	1.17	0.2678	3.7341	-13.880
323K	0.9992	1.19	0.3027	3.3036	-13.994
333K	0.9996	1.20	0.3494	2.8620	-14.030

4. CONCLUSION

Gmelina arborea root extract has been shown to inhibit the corrosion mild steel in 1 M H₂SO₄. Inhibition efficiency increases with an increase in the root extract concentration but decreases with increase in temperature. Physical adsorption of the extract onto mild steel surface has been proposed based on the decrease in inhibition efficiency with increase in temperature, a higher activation energy value in the extract compared to the blank, and the value of free energy of adsorption process being less negative than -20 kJ/mol. The adsorption of *Gmelina arborea* root extract on mild steel surface in 1 M H₂SO₄ has been shown to obey the Langmuir adsorption isotherm.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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