Original Research Article

Inhibition of Mild Steel Corrosion by *Gmelina arborea* Root Extract in 1 M H_2SO_4 solution

.

ABSTRACT

The inhibition of mild steel corrosion in sulphuric acid solution by *Gmelina arborea* root extract has been 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 increases with increase in *Gmelina arborea* root extract concentration but decreases with increase in temperature. The highest inhibition efficiency of 78.59% occurs 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 mild steel surface. The calculated thermodynamic parameters reveal that the adsorption process was endothermic and spontaneous. The phytochemical screening of *Gmelina arborea* root extract reveals the presence of alkaloids, saponins, anthraquinones and terpenes. The adsorption of the root extract on mid steel surface obeys the Langmuir adsorption isotherm.

Keywords: Gmelina arborea, corrosion inhibition, physisorption, extract, Langmuir isotherm

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 sought after 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 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 inhibited 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 H_2SO_4 solution.

2. MATERIALS AND METHODS

2.1Test 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 press - 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 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 H_2SO_4 solution.

2.3 Phytochemical screening of *Gmelina arborea* Root Extract

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 H_2SO_4 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 H_2SO_4 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]:

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

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 the equation [1]:

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

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. 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]:

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

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.

3. RESULTS AND DISCUSSION

3.1 Results of Phytochemical Analysis Of *Gmelina Arborea* Root Extract Ethanol Rind Extract

The results of the phytochemical anlysis of ethanol *Telfairia occidentalis* rind 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 extract concentration of 2.0 g/L at 30°C by the weight loss method. 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 1) 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.

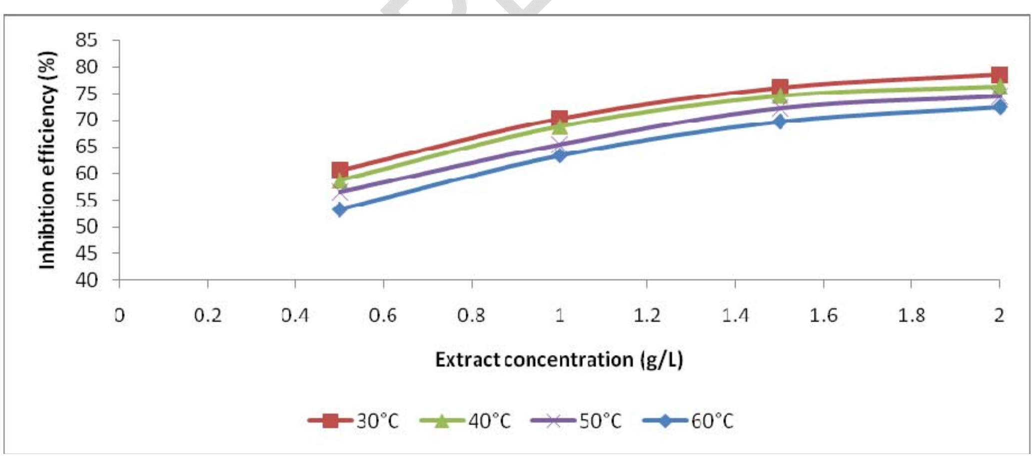

Fig.1. Effect of *Gmelina arborea* root extract concentration (g/L) on the inhibition efficiency (%) of mild steel corrosion in 1 M H_2SO_4 solution at different temperatures

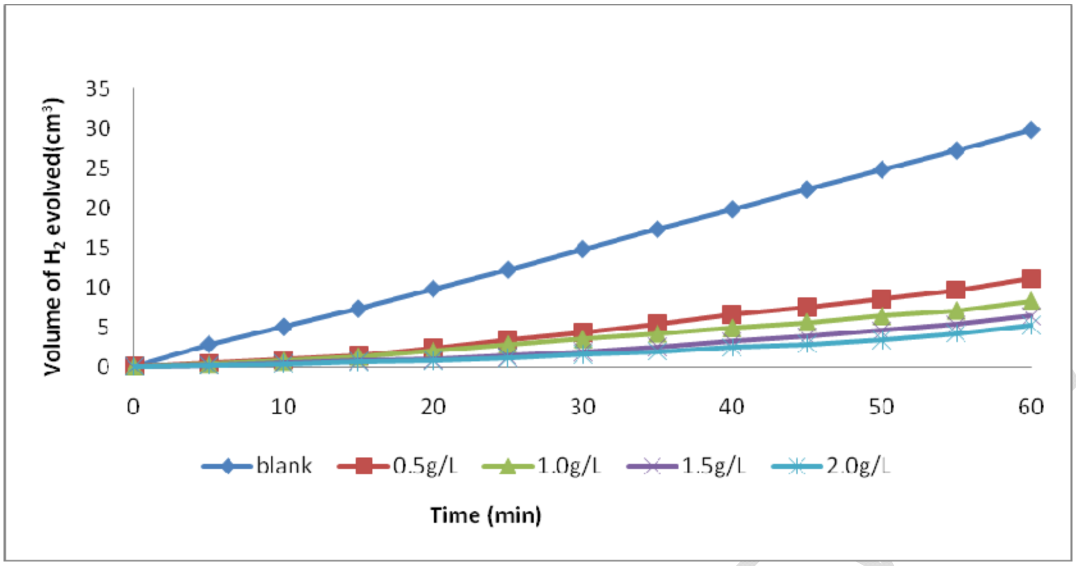

Fig. 2. Variation of volume of H_2 gas evolved (cm^3) with time (min) for mild steel corrosion in 1 M H_2SO_4 in the absence and presence of *Gmelina arborea* root extract at 30°C

Table 1. Effect of *Gmelina arborea* root 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^{-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

3.3 Effect of Temperature on Inhibition Efficiency

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

Table 2. 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

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]:

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

where CR is corrosion rate, R is the universal gas constant, T is the absolute temperature, E_a is activation energy while A is the pre-exponential factor.

Fig 3 illustrates linear plot of In CR vs 1/T for the mild steel corrosion in 1 M H_2SO_4 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 H_2SO_4 – *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 is 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.

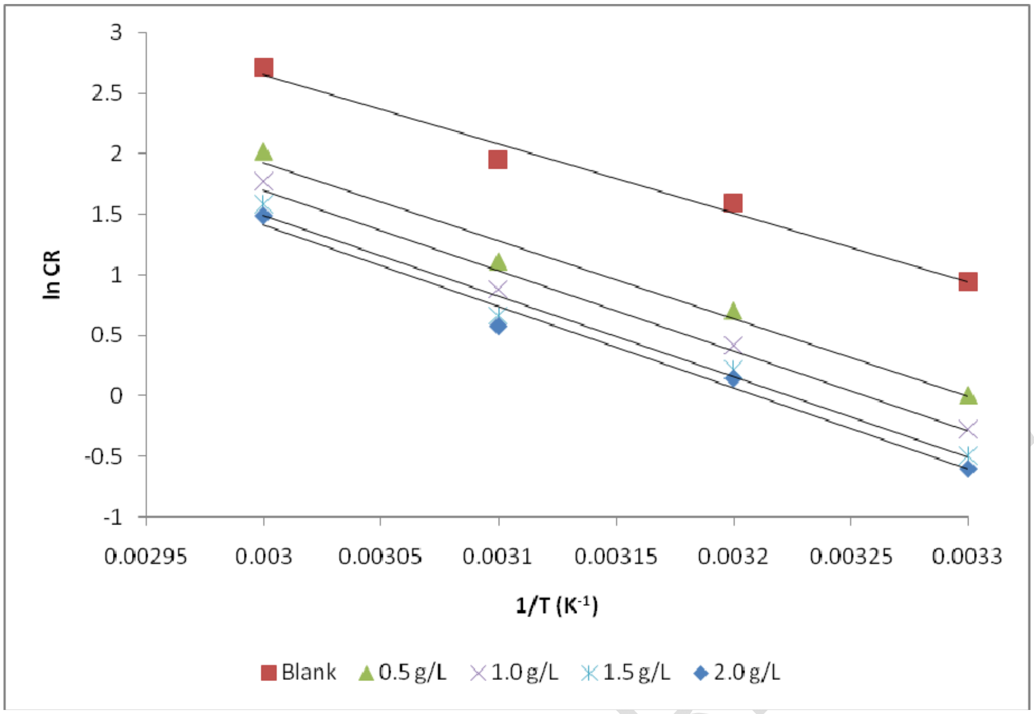

Fig. 3. Arrhenius plot for mild steel corrosion in 1 M H_2SO_4 solution in the presence and absence of *Gmelina arborea* root extract, respectively

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

Extract concentration	E_a (kJ mol^{-1})	ΔH^o_{ads} (kJ mol^{-1})	ΔS^o_{ads} ($JK^{-1}mol^{-1}$)
1 M H_2SO_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

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

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

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 In (CR/T) against (1/T) is shown in Fig 4. This plot has gradient of ($-\Delta H^o_{ads}/R$) and intercept of [ln(R/Nh) + (ΔS^o_{ads} /R)] where the values of ΔS^o_{ads} and ΔH^o_{ads} were obtained. Positive values of ΔH^o_{ads} reveal that the adsorption of *Gmelina arborea* root extract onto mild steel surface was an endothermic process.

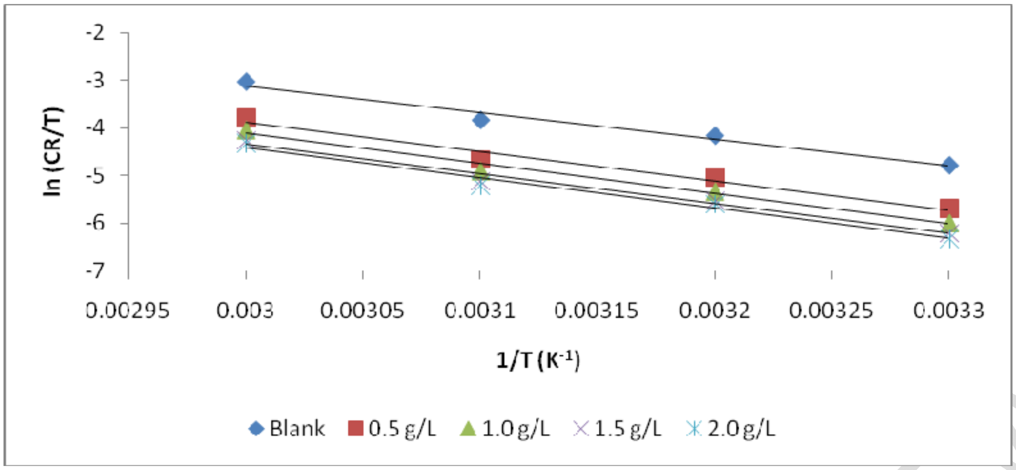

Fig. 4.Transition state plot for mild steel corrosion in 1 M H_2SO_4 solution in the presence and absence of *Gmelina arborea* root extract

3.4 Adsorption Studies

The Langmuir adsorption isotherm proposes that adsorption happens on specific homogenous sites on the metal surface and is used successfully in many monolayer adsorption process [7]. The modified Langmuir isotherm was used to investigate the mechanism of adsorption, which is expressed as [32]:

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

where θ is the degree of surface coverage, C is the inhibition concentration, K_{ads} is the equilibrium adsorption constant. The plot of C/ θ against C (Fig 5) depict a straight line graph supporting that the adsorption of *Gmelina arborea* root extract on mild steel in 1 M H_2SO_4 obeyed the Langmuir adsorption isotherm. The values of K_{ads} which indicate the adsorption strength of the inhibitor onto the metal surface [7], was evaluated from the intercept of the graph. The standard free energy of adsorption ΔG^o_{ads} is determined from the equation [29]:

$$\Delta G^{\circ}_{ads} = -RT\ln(55.5K_{ads}) \qquad (7)$$

where K_{ads} is the equilibrium adsorption constant, ΔG^o_{ads} is the standard free energy of adsorption and the value 55.5 is the molar concentration of water in solution expressed in mol/dm^3

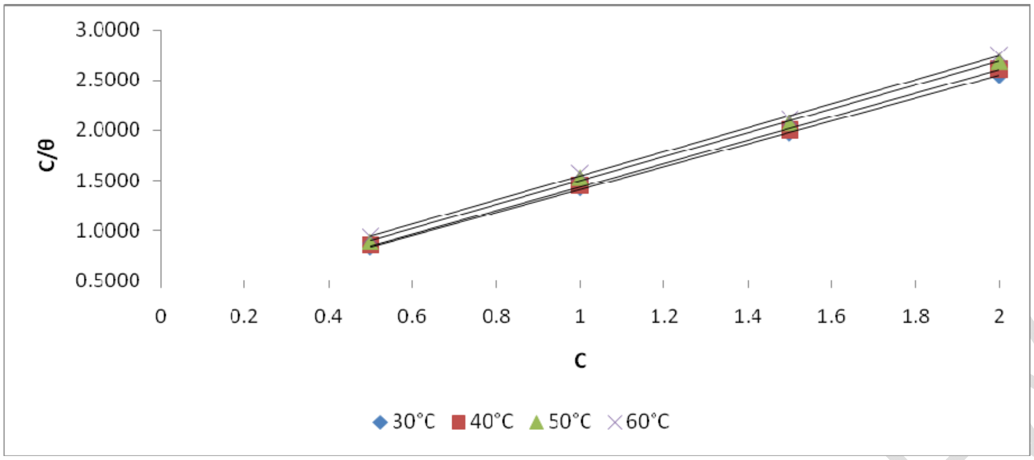

Fig. 5. Langmuir isotherm plot for mild steel corosion in 1 M H_2SO_4 solution containing *Gmelina arborea* root extract at 30°C – 60°C

The values of K_{ads} and ΔG^o_{ads} are represented in Table 4. Values of ΔG^o_{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^o_{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^o_{ads} indicate that the spontaneity of adsorption of *Gmelina arborea* root extract onto the mild steel surface.

Table 4. Some parameters of the linear regression of Langmuir adsorption isotherm for mild steel corrosion in 1 M H_2S0_4 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

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 immersion in 1 M H_2SO_4 solution in the absence of *Gmelina arborea* extract (Fig. 6) 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. 7) reveals a smoother surface. This may be attributed to the formation of an adsorbed film of *Gmelina arborea* molecules on mild steel surface, which inhibit the corrosion of mild steel in 1 M H_2SO_4 solution.

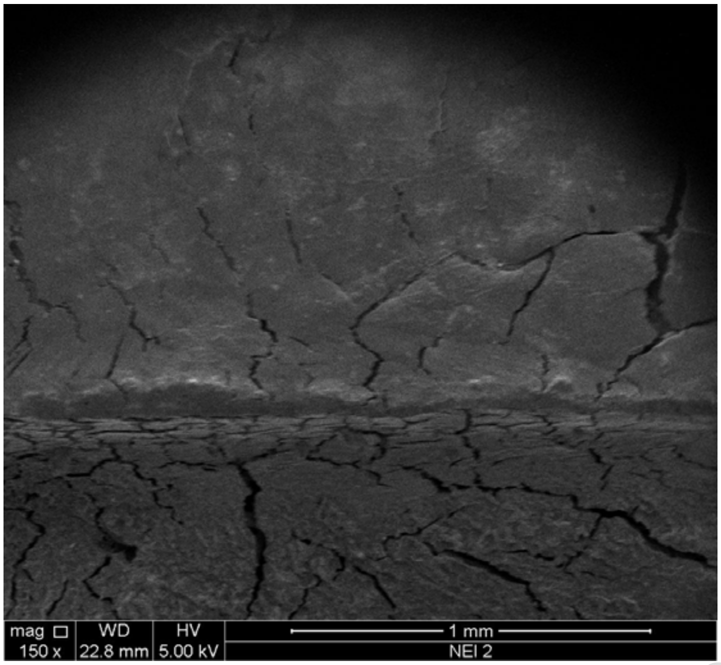

Fig. 6. SEM micrograph of mild steel specimens after 4 hours of immersion in 1 H_2SO_4 solution without *Gmelina arborea* extract

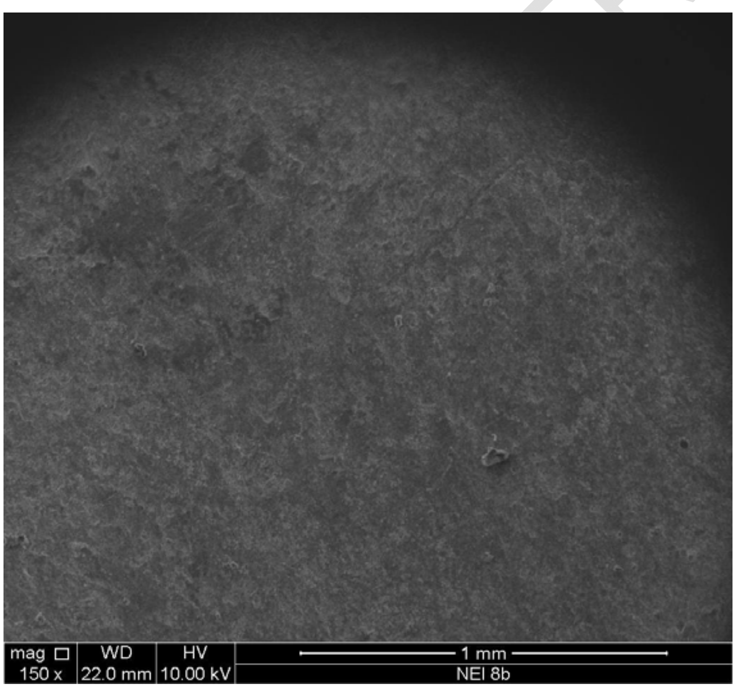

Fig. 7. SEM micrograph of mild steel specimens after 4 hours of immersion in 1 M H_2SO_4 solution containing 2.0 g/L *Gmelina arborea* root extract at 30°C

4. CONCLUSION

Based on the results obtained in this study, the following conclusions could be made: *Gmelina arborea root* extract exhibits good inhibitive action against the corrosion of mild steel in 1 M

H_2SO_4.Inhibition efficiency increases with 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 a decrease in inhibition efficiency with increase in temperature, a higher E_a value in the extract compared to the blank in addition to the ΔG_{ads} of the adsorption process being less negative than -20 kJ/mol. The adsorption of *Gmelina arborea root extract on* mild steel surface in 1 M H_2SO_4 obeys the Langmuir adsorption isotherm.

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