

## Comparative studies of inhibitive properties of *Ficus polita* and *Ficus platyphylla* on corrosion inhibition of mild steel in acidic medium

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**Abstract.** The menace of corrosion of steel in industries has been widely acknowledged. Analysis of oil pipeline failures in oil and gas industries in the Niger Delta area of Nigeria showed corrosion as one of the major causes of failure. Eco-friendly, cheap and renewable materials such as plant extracts have been investigated as alternative to the use of expensive synthetic chemicals which are often hazardous. In this study inhibitive properties of ethanol extracts of *Ficus polita* and *Ficus platyphylla* leaves as eco-friendly inhibitors of mild steel in 2 M sulfuric acid solution were investigated by weight loss method at temperatures of 301, 305, 309 and 313 K. The study has shown that both plant extracts inhibited the corrosion of mild steel in the acid medium. The inhibition efficiency (*IE*) and surface coverage ( $\theta$ ) for both extracts increased with increase in concentration and temperature. This trend is suggestive of chemical adsorption. However, the leaf extract of *Ficus polita* showed higher *IE* compared to *F. platyphylla*. Thermodynamic parameters and apparent activation energy obtained from the studies revealed the inhibition by both plant extracts to be spontaneous, exothermic and chemisorptive, while the adsorption mechanism of both plant extracts on the steel surface aligned with the Freundlich isotherm model, *F. polita* fits well to Temkin and Adejo-Ekwenchi isotherm models in addition.

**Keywords:** steel corrosion; weight loss; thermodynamics; *Ficus polita*; *Ficus platyphylla*; adsorption model.

### 1. Introduction

Corrosion is the main cause of wear in machineries, pipelines and mild steel products in general. One or more forms of corrosion such as pitting, crevices, cracking, fatigue or intergranular are involved during corrosion of mild steel resulting in the failure of components [1]. Corrosion swigs a major part of production cost in industries. The cost of corrosion to the economy of any nation is nefarious [2]. In addition to the cost corrosion adds to environmental pollution which remains a major challenge nations have to deal with [3].

The use of corrosion inhibitors has been considered an easy and cost-effective approach to combating corrosion. Synthetic inhibitors exist and are effective but add to environmental contamination hence alternative and eco-friendly inhibitors are essential [4]. Natural products of plant origin and substances from other renewable sources have gained heightened interest by researchers as green corrosion inhibitors [5-7], because they are biodegradable [8, 9], less toxic [10] and do not contain heavy metals. In the light of these, several plants extracts have been investigated and their corrosion inhibition properties are often attributed to phytochemicals present [11-14]. The active phytochemicals in plants that are effective for corrosion inhibition are those that have heteroatom in their aromatic or long carbon chain [15, 16]. The presence of  $\pi$ -electrons or suitable functional groups from the inhibitor's molecule enables the transfer of charge from

it to the charged metal surface (physical adsorption) or the transfer of electron from the inhibitor's molecule to vacant *d*-orbital of the metal (chemical adsorption) [17-19]. Heterocyclic and phenolic compounds which contains oxygen, sulfur, nitrogen atoms, and multiple bonds in the molecules have active sites for adsorption onto metal surfaces [20, 21] with decrease in efficiency following the order  $P > S > N > O$  [3].

*F. polita* and *F. platyphylla* belong to the family Moraceae, and genus *Ficus*. Several species of *Ficus* have shown excellent percentage inhibition efficiency [22, 23] on mild steel. Usman and coworkers [24] reported tannins, flavonoids, phlobatannins, terpenoids as present in *Ficus polita*. Kubmarawa *et al.* [25] and Akesa and team [26] reported tannins, saponins, flavonoids, glycosides, volatile oils and steroids as present in *Ficus platyphylla*. These tropical plants grow naturally in virgin lands in Benue State and serve mainly for shade in areas where they are planted. This work is aimed at investigation and comparison of the corrosion inhibition efficiency *F. polita* and *F. platyphylla* on mild steel in acidic medium using the weight loss method.

### 2. Experimental

#### 2.1. Materials and reagents

*F. polita* and *F. platyphylla* leaves were obtained from the Main Campus of Benue State University, and identified by the Botanist, in Department of Biological Science, Benue State Makurdi-Nigeria.

Mild steel coupons (3x4 flat bar low carbon mild steel China) were obtained from the Engineering

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The reagents were purchased from Guangdong Guanghua Sci Tech China (absolute ethanol, C<sub>2</sub>H<sub>5</sub>OH 99.7%; sulfuric acid, H<sub>2</sub>SO<sub>4</sub> 98%) and BDH Chemicals Ltd., England (acetone, C<sub>3</sub>H<sub>6</sub>O 99.5%).

## 2.2. Methods

The fresh samples of *F. polita* and *F. platyphylla* leaves were washed to remove dirt and sand particles, shade dried and reduced to powder. 20 g of the sample were extracted by cold maceration in 200 mL of C<sub>2</sub>H<sub>5</sub>OH for 48 hours. The filtrate was separated from the residues over cotton wool and concentrated at 321 K in a thermostat water bath (Clifton, Nickel-Electro Ltd., England) to remove the solvent. The extract was then kept in a desiccator to avoid contamination. Stock solutions of 0.1, 0.2, 0.3, 0.4 and 0.5 g·dm<sup>-3</sup> in 2 M H<sub>2</sub>SO<sub>4</sub> were prepared preceding the analysis. Coupons of dimensions 1.8 cm x 2.0 cm x 0.03 cm were prepared from a sample of mild steel of composition by weight (%) Fe (98.84), Mn (0.70), P (0.04), C (0.18), Si (0.40) and S (0.40). Each coupon was weighed and degreased in acetone, dried and preserved in a desiccator prior to the experiment.

Previously prepared mild steel coupons were suspended without plant extracts (blank) and with plant extracts in varying concentrations of the extracts at varying temperatures (301 to 313 K) in the water bath for 6 hours as reported by Wang *et al.* [18] and Adejo *et al.* [27], with slight modifications. The reaction of each coupon was quenched in a saturated solution of CH<sub>3</sub>COONa, washed in distilled water and dried in acetone and then re-weighed after cooling. The difference in weight was used to calculate the inhibition efficiency and surface coverage ( $\theta$ ) through Equations 1 and 2, respectively [23, 28]:

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

$$\theta = 1 - \frac{W_1}{W_2} \quad (2)$$

where  $W_1$  is weight of the coupon in the absence and  $W_2$  weight of the coupon in the presence of plant extracts, respectively.

## 3. Results and discussion

### 3.1. Inhibition efficiency and surface coverage

The inhibition efficiency and surface coverage were calculated using Equations 1 and 2 respectively by substituting the weight loss data in the absence and presence of the plant extracts. Table 1 and Table 2 show inhibition efficiency and surface coverage of the green corrosion inhibitors on mild steel in 2 M H<sub>2</sub>SO<sub>4</sub> at varying temperatures and concentrations. Inhibition efficiency and surface coverage increased with increase in inhibitor concentration and rise in temperature (K). The increase in IE with rise in temperature is suggestive of chemisorption and consistent with the work reported by [29-31]. As temperature rises, more adsorbate forms chemical bond with the metal surface thereby inhibiting corrosion. *Ficus polita* leaf extract showed higher surface coverage and IE compared to *F. platyphylla* leaf extract. This may be due to the large quantity of phytol

(23.3%); 6, 10, 14-trimethyl-2-pentadecanone (15.0%); (*E*)-6, 10-dimethyl-5, 9-undecadien-2-one (7.3%) and drimenol (5.8%), sesquiterpenes ( $\alpha$ -yiangene,  $\alpha$ -copaene),  $\gamma$ -cadienee, (*E, E*)- $\alpha$ -farnesene in *F. polita* [32]. Although *F. platyphylla* contains volatile oils, terpenoids are absent. Terpenoids are highly conjugated and electron rich compounds which may provide more adsorption sites. This difference in their chemical composition may account for the higher inhibition efficiency observed with *F. polita*. However, reasoning is still open to speculations.

**Table 1.** Variation of IE and  $\theta$  of *Ficus polita* leaf extract with concentration and temperature

Conc. g·dm <sup>-3</sup>	Inhibition efficiency (IE %)				Surface coverage ( $\theta$ )			
	301	305	309	313	301	305	309	313
T [K]								
0.1	17.77	19.13	22.30	27.80	0.1777	0.1913	0.2230	0.2780
0.2	23.42	25.83	27.70	30.04	0.2342	0.2583	0.2770	0.3004
0.3	26.96	28.66	30.95	33.50	0.2696	0.2866	0.3095	0.3350
0.4	36.96	39.21	43.24	46.14	0.3696	0.3921	0.4324	0.4614
0.5	45.82	48.98	53.85	55.96	0.4582	0.4898	0.5385	0.5596

**Table 2.** Variation of IE and  $\theta$  of *F. platyphylla* leaf extract with concentration and temperature

Conc. g·dm <sup>-3</sup>	Inhibition efficiency (IE %)				Surface coverage ( $\theta$ )			
	301	305	309	313	301	305	309	313
T [K]								
0.1	1.65	3.13	6.22	8.41	0.0165	0.0313	0.0622	0.0841
0.2	3.31	5.00	10.52	13.38	0.0331	0.0500	0.1052	0.1338
0.3	6.43	10.83	12.59	18.03	0.0643	0.1083	0.1259	0.1803
0.4	13.60	18.96	19.26	23.57	0.1360	0.1896	0.1926	0.2357
0.5	23.71	25.00	30.74	35.10	0.2371	0.2500	0.3074	0.3510

### 3.2. Calculated values of activation energy and heat of adsorption

The apparent activation energy ( $E_a$ ) was calculated from the Arrhenius equation given as [22, 33]:

$$\log C_R = \log A - \left( \frac{E_a}{2.303RT} \right) \quad (3)$$

$E_a$  was obtained from the slope of the plot of  $\log C_R$  against  $1/T$ , where  $C_R$  is corrosion rate (mg/cm<sup>2</sup>·h),  $A$  is the constant frequency factor,  $E_a$  is the apparent activation energy,  $R$  is molar gas constant and  $T$  is absolute temperature. The values of heat of adsorption ( $Q_{ads}$ ) were obtained from the slope  $Q_{ads}/2.303R$  of the plot of  $\log \theta/1-\theta$  against  $1/T$ .

Tables 3 and 4 show the data for activation energy ( $E_a$ ) and thermodynamic properties ( $Q_{ads}$ ,  $\Delta H$ ,  $\Delta S$  and  $\Delta G$ ) for the process of corrosion inhibition of extracts of *Ficus polita* and *Ficus platyphylla* respectively. The apparent activation energy ( $E_a$ ) of the inhibition process decreased with increasing inhibitor concentration. Values of activation energy in physisorption process are lower than 40 kJ/mol, while higher than this limit is chemisorption [33]. This implies that the green inhibitors are adsorbed to the surface of the metal by chemical bonding.

### 3.3. Thermodynamic properties

The enthalpies ( $\Delta H$ ) of adsorption of both plant extracts decreased with increase in inhibitor concentration, the strength of the adsorption increased with increase in the concentration of the inhibitor indicative of interaction between the metal surface and the inhibitors [27]. The values of heat of adsorption for both *F. polita* and *F. platyphylla* extracts were positive implying that both

processes are endothermic and did take up energy from the surrounding. The Gibb's free energy is negative implying spontaneity of the adsorption reactions. It is also important to note that values of Gibb's free energy of adsorption, greater than  $-40$  kJ/mol are consistent with the transfer of electron from the inhibitor to the metal surface which represents a chemical adsorption whereas, values of Gibb's free energy less than  $-40$  kJ/mol signifies the mechanism of physical adsorption [34]. This confirms further the mechanism of adsorption of both green inhibitors is chemisorption as the values of the Gibb's free energy are greater than  $-40$  kJ/mol.

**Table 3.** Activation energy ( $E_a$ ) and thermodynamic parameters ( $\Delta H$ ,  $\Delta S$  and  $\Delta G_{ads}$ ) of *F. polita* leaf extract inhibition process

Conc. g·dm <sup>-3</sup>	$E_a$ (kJ/mol)	$\Delta H$ (kJ/mol)	$\Delta S$ (J/K)	$-Q_{ads}$ (kJ/mol)	$\Delta G_{ads}$ (kJ/mol)
Blank	75.54	73.04	209.38	-	301 305 309 313
0.1	70.30	67.79	366.24	25.24	21.72 24.14 29.61 40.24
0.2	70.45	67.94	366.08	18.80	15.56 17.73 19.77 22.44
0.3	70.06	67.55	364.41	18.04	12.37 13.64 15.42 17.55
0.4	65.75	63.24	348.87	42.14	14.73 16.42 19.65 22.38
0.5	62.35	59.84	336.31	25.74	17.00 19.55 24.08 26.56

**Table 4.** Activation energy and thermodynamic parameters ( $Q_{ads}$ ,  $\Delta H$ ,  $\Delta S$  and  $\Delta G_{ads}$ ) of *F. platyphylla* leaf extract inhibition process

Conc. g·dm <sup>-3</sup>	$E_a$ (kJ/mol)	$\Delta H$ (kJ/mol)	$\Delta S$ (J/K)	$-Q_{ads}$ (kJ/mol)	$\Delta G_{ads}$ (kJ/mol)
Blank	67.19	66.60	362.00	-	301 305 309 313
0.1	62.18	61.79	346.39	117.19	1.686 3.290 6.843 9.597
0.2	59.40	56.91	329.33	106.28	1.720 2.680 6.065 8.072
0.3	58.83	56.32	327.00	73.06	2.302 4.123 4.953 7.663
0.4	60.14	57.63	330.66	39.33	3.955 5.956 6.153 8.057
0.5	55.98	54.40	323.70	38.77	6.247 6.789 9.159 11.305

### 3.4. Adsorption isotherms

The linearized forms of the isotherms studied are given in the equations below [35, 36]:

$$\text{Langmuir: } \frac{C}{\theta} = \frac{1}{K} + C \quad (4)$$

$$\text{Freundlich: } \log \theta = \log K_F + n_F \log C \quad (5)$$

$$\text{Temkin: } \frac{-2\alpha\theta}{2.303} = \log k + \log C \quad (6)$$

$$\text{El-Awady: } \log \frac{\theta}{(1-\theta)} = \log K + y \log [C] \quad (7)$$

$$\text{Adejo- Ekenchi: } \log \frac{1}{1-\theta} = \log K_{AE} + b \log C \quad (8)$$

where  $\theta$  is surface coverage,  $K$  is corrosion rate constant,  $n_F$  is Freundlich constant, the parameter  $\alpha$  indicates the type of interaction at adsorbent surface,  $y$  is number of water molecules replaced by corrosion inhibitor in El-Awady isotherm,  $b$  is slope, and  $K_{AE}$  is the Adejo-Ekwenchi adsorption constant.

The data for adsorption isotherms shown in Table 5 for Langmuir, Freundlich, El-Awady, Temkin and Adejo-Ekwenchi isotherms were employed to study the adsorption mechanism of *F. polita*. The values of the equilibrium constants ( $K_{ads}$ ) were all positive, indicative of favorable adsorption. The  $R^2$  values of Langmuir isotherm is a bit far less than unity thus the adsorption cannot be modeled by this isotherm. The  $R^2$  values in Freundlich isotherm are close to unity and also the

values of  $n$  parameter are close to the typical value of 0.6 [27] and thus these data can be modeled by this isotherm. The fitting of the adsorption into Temkin isotherm model confirms further the chemical adsorption mechanism of the inhibitor onto the metal surface. The negative values of the  $\alpha$  parameter is indicative of repulsive interaction of the molecules within the layer of adsorption. Temkin isotherm model fitting is a feature of chemisorption [37]. For El-Awady isotherm, values of  $y > 1$  implies a multilayer formation and values of  $y < 1$  implies monolayer, that is an inhibitor occupies more than one active site. The values of  $y$  from this study were less than one indicative of mono layer formation and confirming further chemisorption adsorption mechanism [27, 38]. The  $b$  parameter in the Adejo-Ekwenchi isotherm is useful in resolving the mechanism of adsorption. Values of parameter  $b$  decreasing with rise in temperature shows physical adsorption and increase or fairly constant  $b$  value with increase in temperature indicates chemisorption. The value of  $b$  in this study increases with increase in temperature which signifies chemical adsorption mechanism [39].

**Table 5.** Adsorption isotherm parameters for adsorption of *F. polita* leaf extract onto the mild steel surface

Isotherm	$T$ [K]	$R^2$	Slope	Intercept	$K_{ads}$	$-\Delta G_{ads}$
Langmuir	301	0.7741	1.2666	0.5410	1.8484	11.5889
	305	0.7486	1.2420	0.5043	1.9829	11.9206
	309	0.7103	1.1633	0.4497	2.2237	12.3713
	313	0.7710	1.1513	0.4023	2.4857	12.8213
Freundlich	301	0.9484	0.5749	-0.1999	0.6311	8.8992
	305	0.9314	0.5561	-0.1883	0.6482	9.0855
	309	0.8934	0.5247	-0.1638	0.6858	9.3495
	313	0.9114	0.5102	-0.1431	0.7193	9.5944
Temkin	301	0.8739	0.3830	0.5319	1.7022	11.3830
	305	0.8531	0.3900	0.5538	1.7399	11.5893
	309	0.8169	0.4180	0.6004	1.8228	11.8607
	313	0.8407	0.4300	0.6305	1.8785	12.0926
El-Awady	301	0.9201	0.8192	0.1076	1.3532	10.8082
	305	0.8979	0.8146	0.1382	1.4780	11.1754
	309	0.8529	0.8167	0.2050	1.2859	10.9644
	313	0.8708	0.8207	0.2530	2.0365	12.3026
Adejo-Ekwenchi	301	0.8343	0.2443	0.3074	2.0296	11.8227
	305	0.8096	0.2585	0.3265	2.1208	12.0911
	309	0.7701	0.2921	0.3692	2.3399	12.5021
	313	0.7926	0.3106	0.3967	2.4929	12.8288

Table 6 shows the fitting of data into various adsorption isotherms for the *F. platyphylla* leaf extract onto the mild steel surface. The  $R^2$  values show that the adsorption data fit into Freundlich isotherm. The equilibrium constants ( $K_{ads}$ ) were positive implying favorable adsorption. Although the  $R^2$  values of Freundlich isotherm are close to unity, the  $n$  parameter is greater than 0.6 [40]. The  $y$  values for El-Awady isotherm are greater than 1 which implies the reaction forms multilayer suggesting physical adsorption

mechanism, hence the data does not fit into this isotherm.

**Table 6.** Adsorption isotherm parameters for adsorption of *F. platyphylla* leaf extract onto the mild steel surface

Isotherm	T [K]	R <sup>2</sup>	Slope	Intercept	K <sub>ads</sub>	-G <sub>ads</sub>
<i>n</i>						
Freundlich	301	0.9544	1.6407	0.2305	1.7002	1.6407 11.38
	305	0.9552	1.3484	0.2313	1.7033	1.3484 11.54
	309	0.9309	0.9195	0.3251	2.1140	0.9195 12.24
	313	0.9598	0.8107	0.2731	1.8754	0.8107 12.09
<i>-α</i>						
Temkin	301	0.7362	0.1223	0.2619	1.2994	0.0612 10.71
	305	0.8408	0.1339	0.3060	1.3580	0.0670 10.96
	309	0.7718	0.1320	0.3363	1.3998	0.0660 11.18
	313	0.8323	0.1461	0.3960	1.4859	0.0731 11.48
<i>y</i>						
El-Awady	301	0.9419	1.7793	0.1032	1.1429	10.39
	305	0.9474	1.5031	0.0805	1.1312	10.50
	309	0.9077	1.0799	0.1540	1.3887	11.16
	313	0.9358	0.9963	0.0650	1.1620	10.84

#### 4. Conclusions

Ethanol leaf extracts of *F. polita* and *F. platyphylla* were investigated for their corrosion inhibition properties onto the surface of mild steel in acidic medium, with *F. polita* showing higher inhibition efficiency than *F. platyphylla*. The chemisorption adsorption mechanism of *F. polita* best fit Freundlich, Temkin and Adejo-Ekwenchi isotherm models, while chemisorption mechanism of *F. platyphylla* fits well Freundlich adsorption isotherm model.

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#### Conflict of interest

Authors declare no conflict of interest.

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