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Evaluation of *Barleria prionitis* Leaves Extract as A Green Corrosion Inhibitor for Mild Steel in Acid Medium

A. Mushira banu[1]*and K. Riaz Ahamed[1]
[1]PG and Research Department of chemistry, Jamal Mohamed college, Tiruchirappalli-20,

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Corresponding Author Email: mushira330@gmail.com

Abstract
The corrosion protection of mild steel specimens immersed in 1.0 M H_2SO_4 by *Barleria prionitis* leaves extract was investigated by gravimetric, electrochemical techniques (potentiodynamic polarization and electrochemical impedance spectroscopy). The potentiodynamic polarization results revealed that the aqueous extract of *Barleria prionitis* act as a mixed type of inhibitor. The inhibiting effect was studying using impedance measurements showed that the double-layer capacitance decreased with increase in the inhibitor concentration. The mechanism of adsorption of *Barleria prionitis* leaves extract on mild steel surface was analysed by using various adsorption isotherms. To estimate the corrosion inhibition potential of *Barleria prionitis* in acid medium on mild steel (MS) with a view to develop green corrosion inhibitors. This green inhibitor can find use in the inhibition of corrosion in various industries. The results indicate that leaves of *Barleria prionitis* serve as a good eco-friendly corrosion inhibitor.

Keywords
Barleria prionitis, Mild steel, Corrosion, Weight loss, Adsorption isotherms, Polarization, Impedance.

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INTRODUCTION

Corrosion is the critical phenomenon which distresses almost all metals. Iron is the most widely used metal and one of the first for which corrosion was met. Corrosion inhibitors are one of the superlative approaches to control corrosion. Ecological concerns necessitate corrosion inhibitors to survey certain guidelines. Most of the conservative inhibitors that have been industrialized till today are non-biodegradable and highly contaminated to human beings [1]. In recent years, plant extracts have become the attention of corrosion inhibition research due to their low toxicity, easy and cost-effective preparation. Extracts of various leaves like *Wrightiatinctoria, Clerodendrumphlomidis, Ipomoeatriloba* was investigated by N.S. Patel *et al*.[2]. An evaluation of effective performance of *Citrus aurantium* leaves extract on the corrosion of mild steel in 1 M H_2SO_4 was carried out by Karim H. Hassan *et al*.[3]. S. Jyothi and J. Ravichandran[4] observed inhibitive effects of acid extract of *coccinia indica* leaves on the corrosion of mild steel. various plant extracts such as Rubber leaf, Houttuynia Cordata extract, *Pennisetum purpureum* extract, *Capsicum annuum* fruit paste, to prevent corrosion of metals were also studied [5-18]. Hence in the present work it has been projected to gain some insight into the corrosion of mild steel in presence of *Barleria prionitis* leaves extract as a corrosion inhibitor. Weight loss method and electrochemical methods such as polarization study and AC impedance spectra have been used.

MATERIALS AND METHODS

The investigational studies connected to the inhibition action of *Barleria prionitis* leaves extract on mild steel in 1.0 M H_2SO_4 have been offered here. Figure 1 shows the leaves of *Barleria prionitis*. The leaves were collected and authenticated by Dr. John Britto, The Rapient Herbarium and Centre for Moduler Systematics, St.Joseph's college, Trichirappallli, Tamilnadu, India.

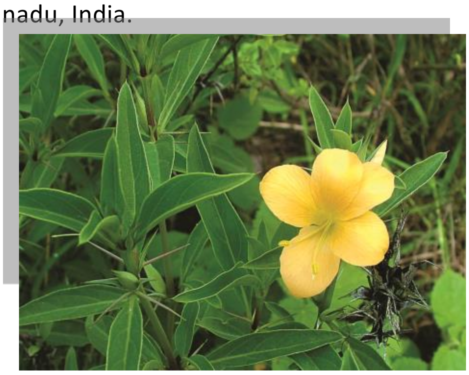

Figure 1: *Barleria prionitis* plant

The botanical study of *Barleria prionitis* as follows: **Botanical name:** *Barleria prionitis*; **Tamil name:** Sullimalar, **Kingdom :** Plantae**; Order** : Lamiales**; Family** : Acanthaaceae**; Genus** : *Barleria***; Species** : *B. prionitis;* **Common Names :** Porcupine flower

Its leaves are known to contain 6-Hydroxyflavone, the major chemical compound that is a noncompetitive inhibitor of the protein cytochrome P450 2C9[19].

Preparation of extract

The fresh leaves of *Barleria prionitis* were collected and dried in shade and then crumbled to powder. *Barleria prionitis* powder (10g) was boiled with 100 ml double distilled water and condensed to 50 ml. The extract was left to cool down and then filtered using Whatman filter paper. From that various concentrations were prepared [20].

Materials and chemicals used

Mild steel was employed as test material for this research. For polarization, cylindrical mild steel rod entrenched in Teflon leaving a working area of 1.0 cm^2. The electrodes were refined with emery papers of 0/0, 2/0, 3/0 and 4/0 grades and degreased with acetone, dried and used.

Weight loss method

The polished and pre-weighed mild steel samples were tied with threads and suspended in 1.0 M H_2SO_4 test solutions, with and without the inhibitor of various concentrations. After the specimens were cautiously washed in double-distilled water, dried and then weighed. The Corrosion rate and inhibition efficiency (I.E.%) of the extract were calculated using the equations given below [21],

Corrosion Rate (mpy)= 87.6×W/ADT

W= Weight loss in mg, A=Area of the mild steel sample in cm^2, D=Density of the mild steel g/cm^3; T= Exposure time in hrs.

Inhibition efficiency (%) = W_0-W_i/W_0 × 100

Where, W_0=Weight loss in blank H_2SO_4, W_i=Weight loss in the presence of leaves extract

Electrochemical studies

The electrochemical studies were completed with a three-electrode cell assembly. The middle neck was used to accommodate working electrode (WE) of mild steel and the remaining two for the reference electrode (RE/SCE), and the counter electrode (CE) of platinum (99.5% pure) [22].

Electrochemical methods of corrosion testing concerning linear polarization and Tafel plots using sophisticated potentiostate devices give a rapid corrosion rate which tracks the type of inhibition as anodic, cathodic or mixed type. The electrochemical impedance study verifies the adsorption of phytochemical constituents present in the leaves extract by increase in double layer capacitance values from blank H_2SO_4 solution. A solution quantity of 100 ml of the test solution was taken in a polarization cell. The working electrode was polished with 0/0, 1/0, 2/0, 3/0, and 4/0 emery papers consecutively and degreased with acetone, the working electrode, reference and auxiliary platinum electrodes were gathered, and connections were made. Stirring was provided to the test solutions to avoid for the system the concentrations polarization before start of experiment. A time interval of about 15 minutes was given for the system to attain state and open circuit potential was noted.

RESULTS AND DISCUSSION

Weight loss study

In order to determine the corrosion rate (C.R) and percentage of inhibition efficiency (% I.E.) by weight loss experiments were performed for mild steel in 1.0 M H_2SO_4 with various concentrations of *Barleria prionitis* extract ranging from 0.2 % to 1.0 %. The corresponding values of inhibition efficiency and corrosion rate for three different temperatures are given in Table 1. It is perceived that as the concentration of inhibitor increases, inhibition efficiency also increases, and corrosion rate decreases which show the maximum inhibition efficiency of 92.15% for 1.0 % *Barleria prionitis* leaves extract.

Effect of temperature:

In electrochemical reaction, the corrosion rate of metal almost doubles for every 10 °C rise in temperature. Table 1 inferred that the temperature increases from room temperature, corrosion rate also increases. The inhibition efficiency values are decreases to 73.46 % at 323 K. It indicates that the desorption process of protective layer from the metal surface.

Table 1: Inhibition effect on corrosion of mild steel in 1.0 M H_2SO_4 by *Barleria prionitis* leaves extract at various temperatures

System	Inhibitor Concentration, V/V (%)	303 K C. R (mpy)	303 K I.E. (%)	313 K C. R (mpy)	313 K I.E. (%)	323 K C. R (mpy)	323 K I.E. (%)
BP-H_2SO_4	Blank	142.0992	-	252.1565	-	362.21	-
	0.2	48.75954	65.68	133.7405	52.48	211.75	41.53
	0.4	33.43511	76.47	90.55344	64.08	186.67	48.46
	0.6	25.07634	82.35	65.47710	74.03	162.99	55.00
	0.8	19.50382	86.27	45.97328	81.76	136.52	62.30
	1.0	11.14504	**92.15**	33.43511	**86.74**	96.120	**73.46**

Adsorption isotherms

The phenomenon of interface between the metal surface and inhibitor can be better understood in relations of adsorption isotherm. The interaction mode and degree of surface coverage between an inhibitor and a metallic surface plays an significant role to understand this part of the study [23]. The adsorption isotherms were performed by using the experimental data which obtained from weight loss measurements. Langmuir, Temkin adsorption isotherms were applied. Various adsorption parameters are tabulated in tables 2 and 3 and the corresponding graphs are shown in Figs. (2,3).

Langmuir adsorption isotherm

The dependence of fraction of the surface covered (θ) obtained by the ratios (I.E.%/100) as a function of the extract concentration (C) was graphically showed in Langmuir adsorption isotherm.

$$C/\theta = 1/K_{ads} + C$$

Where, C is the inhibitor concentration; θ is the fraction of the surface covered, K_{ads} is the adsorption coefficient.

Table 2: Langmuir adsorption isotherm for the inhibition of corrosion of mild steel in 1.0 M H_2SO_4 using *Barleria prionitis* leaves extract

C, (%)	C/Θ-303 K	C/Θ-313 K	C/Θ-323 K
0.2	0.304507	0.381098	0.48158
0.4	0.523081	0.624220	0.825423
0.6	0.728597	0.810482	1.090909
0.8	0.927321	0.978474	1.284109
1.0	1.085187	1.152871	1.361285

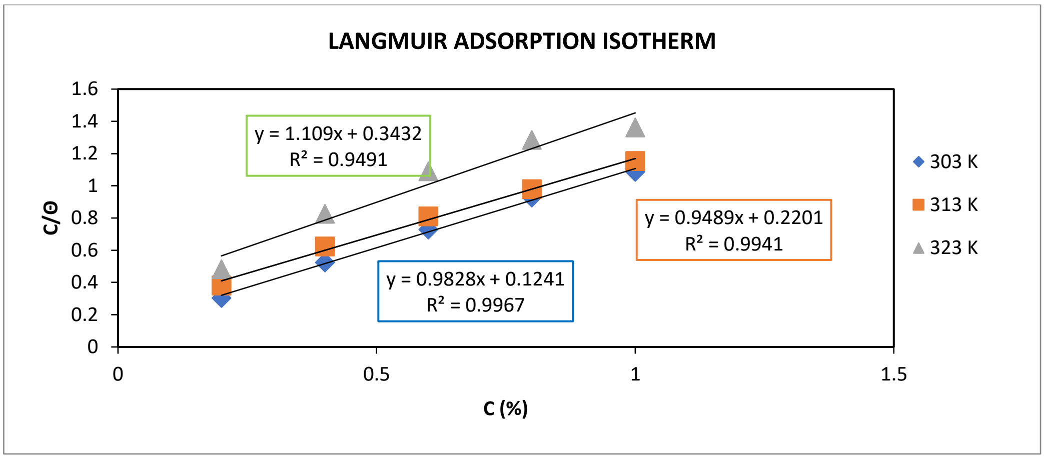

Figure 2: Langmuir adsorption isotherm for the inhibition of corrosion of mild steel in 1.0 M H_2SO_4 using *Barleria prionitis* leaves extract at various temperatures

Temkin adsorption isotherm

According to Temkin adsorption isotherm, the degree of surface coverage (θ) is correlated to inhibitor concentration (C) according to expression

Exp (-2aθ) = KC

Where K is the adsorption equilibrium constant and a, is the attractive parameter. Rearranging and taking logarithm of both sides

θ = -2.303 log K/ 2a – 2.303 log C /2a

Table 3: Temkin adsorption isotherm for the inhibition of corrosion of mild steel in 1.0 M H_2SO_4 using *Barleria prionitis* leaves extract

2+log C	Θ-303 K	Θ-313 K	Θ-323 K
1.3010	0.6568	0.5248	0.4153
1.6020	0.7647	0.6408	0.4846
1.7781	0.8235	0.7403	0.5500
1.9030	0.8627	0.8176	0.62300
2.0000	0.9215	0.8674	0.7346

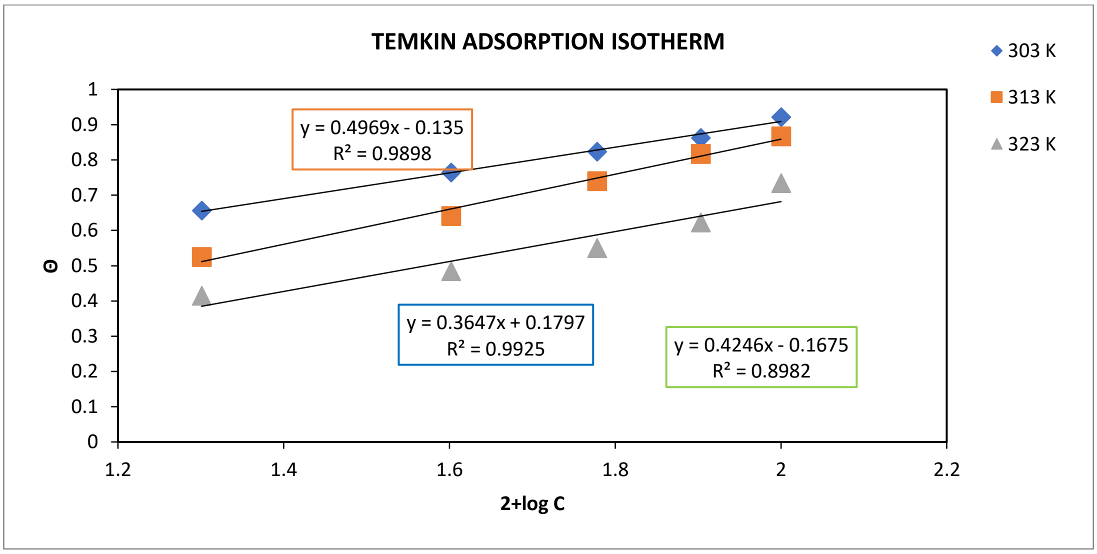

Figure 3: Temkin adsorption isotherm for the inhibition of corrosion of mild steel in 1.0 M H_2SO_4 using *Barleria prionitis* leaves extract at various temperatures

Plot of θ against 2+log C contributed a linear relationship demonstrating that Temkin adsorption isotherm was obeyed.

The $\mathbf{R^2}$ values show that Langmuir as well as Temkin is best fit isotherms. The values show that the plot has good correlation coefficient and linearity. The R^2 values are very close to unity, indicating strong commitment to both adsorption isotherms.

Potentiodynamic polarization studies

Polarization study has been used to examine the protective layer formation during the process of corrosion inhibition study. Corrosion current value (I_{corr}) decreases, when there is corrosion inhibition [24].

Figure 4 shows the cathodic and anodic Tafel polarization curves of mild steel in 1.0 M H_2SO_4 without and with different concentrations of *Barleria prionitis* leaves extract. Tafel extrapolations of the anodic and cathodic lines of the polarization curves were used to determine various electrochemical parameters such as anodic and cathodic Tafel slopes (b_a and b_c), potential (E_{corr}), and current density of corrosion (I_{corr}) as given in Table 4.

Table 4 displays a decrease in corrosion current density (I_{corr}) with the addition of different concentration of the inhibitor.

Table 4: Tafel polarization parameters obtained from polarization curves for mild steel in 1.0 M H_2SO_4 in the absence and presence of inhibitor

System	Concentration of inhibitor, % (v/v)	$-E_{corr}$, mV/SCE	I_{corr}, A/cm^2	b_c, mV/decade	b_a, mV/decade	LPR	% I.E.
BP-Leaves extract	Blank	0.473	3.103×10^{-3}	6.032	10.29	8.60	-
	0.2	0.465	9.853×10^{-4}	135.2	93.90	24.5	**68.24**
	1.0	0.465	3.165×10^{-4}	161.4	80.00	70.0	**89.80**

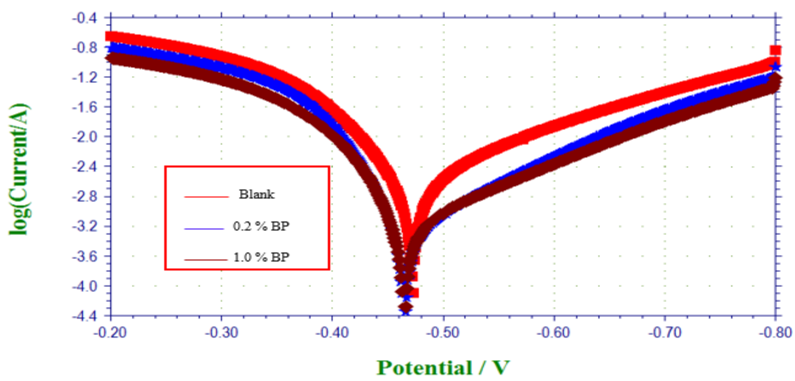

Figure 4: Polarization curves for 1.0 M H_2SO_4 in the absence and presence of *Barleria prionitis* leaves extract

AC impedance spectra

The experimental results attained from the AC impedance spectra for the corrosion of mild steel in the absence and presence of the inhibitor are summarized in Table 5. The impedance spectra for the mild steel speciemens in 1.0 M H_2SO_4 in the absence of *Barleria prionitis* leaves extract and in the presence of various concentrations of *Barleria prionitis* leaves extract are presented in Fig 5. An extensive increase in the total impedance was observed with the addition of *Barleria prionitis* leaves extract. The double layer capacitance (C_{dl}) values are decreases and charge transfer resistance values (R_{ct}) are increases with the concentrations of the inhibitor. From the observation the protective layer formation due to the active components adsorbed on the surface of the mild steel present in the plant extract was confirmed [25]. C_{dl} was calculated by using the following expression,

$$C_{dl} = \frac{1}{2\,\pi\, Rct\, fmax}$$

$\mathbf{f}_{max}$ = Maximum frequency, R_{ct} = charge transfer resistance.

Table 5: Impedance parameters for mild steel in 1.0 M H_2SO_4 in the absence and presence of *Barleria prionitis* leaves extract

System	Concentration of inhibitor, % (v/v)	R_{ct}, Ω	C_{dl}, μF/cm^2	Imp	Ph	I.E. (%)
	Blank – H_2SO_4	2.1510	7.657×10^{-7}	0.5337	27.37	-
BP -Leaves extract	0.2	6.4115	2.568×10^{-7}	1.2252	57.10	**66.46**
	1.0	8.6380	1.906×10^{-7}	1.2909	61.16	**74.77**

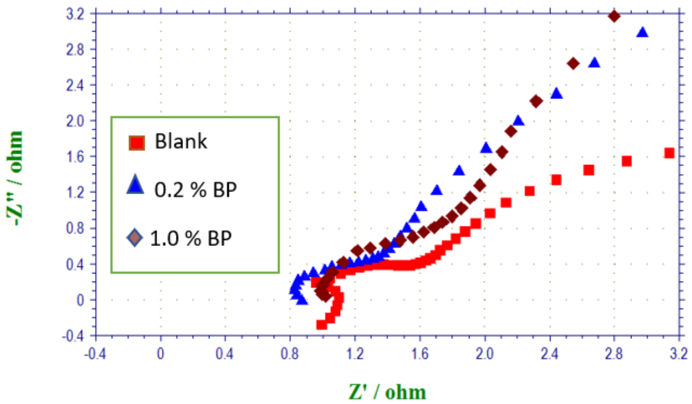

Figure 5: Impedance curves for mild steel in 1.0 M H_2SO_4 in the absence and presence of *Barleria prionitis* leaves extract

Table 6 predicts the comparison of inhibition efficiencies of three independent methods. It shows the close agreement.

Table 6: Comparison of inhibition efficiencies measured by weight loss, polarization and impedance studies for 1.0 M H_2SO_4

[Inhibitor], (%)	Inhibition Efficiency (%)		
	Weight loss studies	Polarization studies	Impedance studies
0.2	67.68	68.24	66.46
1.0	92.15	89.80	74.77

CONCLUSION

The results obtained from the weight loss method demonstrated that the *Barleria prionitis* extract acts as an effective inhibitor on mild steel corrosion in 1.0 M H_2SO_4.

Polarization curves established that the *Barleria prionitis* is a mixed-type inhibitor for mild steel surface corrosion in these solutions.

The values obtained from the weight loss technique for the studied inhibitor fit into the Langmuir and Temkin adsorption isotherms.

The corrosion process was inhibited by adsorption of the active components on the mild steel surface.

The inhibition efficiency was increased with the increase of the *Barleria prionitis* Concentrations and decreases at high temperatures.

The values of inhibition efficiencies obtained from the different independent techniques showed the validity of the obtained results. Thus the *Barleria prionitis* leaves extract was proved to be an effective eco-friendly and inexpensive inhibitor.

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