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Research Article

CORROSION MITIGATION OF MILD STEEL IN ACID MEDIA BY *WATTAKAKA VOLUBILIS* LEAVES EXTRACT

Mushira Banu A* and Riaz Ahamed K

PG and Research Department of Chemistry, Jamal Mohamed College (Autonomous), Tiruchirappalli, India

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ABSTRACT

The corrosion mitigation of mild steel in 1.0 N HCl by *Wattakaka volubilis* leaves extract have been studied at room temperature using weight loss method, potentiodynamic polarization and electro-chemical impedance spectroscopy techniques. The results show that the inhibition efficiency increases with the increase of the extract concentration. The adsorption of the phyto-chemical components present in the leaves extract on the mild steel surface obeys Langmuir adsorption isotherm and occurs spontaneously. The thermodynamic parameters for the adsorption process were calculated. These thermodynamic parameters show strong interaction between inhibitor and mild steel surface.

INTRODUCTION

Corrosion of metals is a spontaneous natural process. To control corrosion rate in cooling water systems many inhibitors are used. To avoid environmental toxicity, extract of natural products are used. For this purpose, natural products are used by many researchers.

The use of plant extracts as corrosion inhibitors has gained prominence as replacement for synthetic organic compounds. The plant natural products have been found to be effective, cheap and eco-friendly anticorrosion agents. Corrosion inhibitions of essential oils of Alpinia galanga were investigated on mild steel in hydrochloric acid solution using weight loss method by Ajeigbe *et al*.(2017). Sanaei *et al*.(2017) have used an effective green corrosion inhibitive hybrid pigment based on zinc acetate-Cichorium intybus L leaves extract to control corrosion of mild steel in aqueous chloride solutions.

A comparative study on the inhibitory action of some green inhibitors on the corrosion of mild steel in hydrochloric acid medium has been reported by Shyamala and Arulanantham (2017). The use of morinda citrifolia as a green corrosion inhibitor for low carbon steel in 3.5% NaCl solution has been by Kusumastuti *et al*. (2017). Evaluation of Thymus vulgaris plant extract as an eco-friendly corrosion inhibitor for stainless steel 304 in acidic solution by means of electrochemical studies has been done by Ehsani *et al*.(2017). They have used density functional theory also. Rajendran *et al*.(2004 & 2005) and Sangeetha *et al*.(2011) have used extracts of various plant materials such as henna leaves, curcumin, caffeine, spirulina, extracts to control of corrosion of metals.

The present work is undertaken to evaluate the inhibition efficiency of an aqueous extract of *Wattakaka volubilis* leaves in controlling corrosion of mild steel in HCl medium. Weight loss method and electrochemical methods such as polarization study and AC impedance spectra have been used.

Experimental Section

Wattakaka Volubilis

Fresh leaves of *Wattakaka volubilis* were collected from cuddalore district, aqueous extract is prepared and used for present study. The plant picture is shown in Fig. 1. The leaves were authenticated and identified by Dr. John Britto, The Rapient Herbarium and Centre for Moduler Systematics, St.Joseph's college, Trichirappallli, Tamilnadu, India.

Corresponding author:* **Riaz Ahamed K
Department of Chemistry, Jamal Mohamed College (Autonomous), Tiruchirappalli, India

Fig 1 *Wattakaka Volubilis* Plant

Botanical Study

Botanical name: *Wattakaka volubilis*; Tamil name: Kodi Palai, Kurinja*;* Kingdom: Plantea*;* Order: Gentianales; Family: Asclepiadacea;Genus: Wattakaka; Species: W.volubilis

Phyto-chemical screening of Wattakaka volubilis

Shibu and Dhanam 2014, gives the phyto-chemical screening of *Wattakaka volubilis* leaves extract in aqueous solvent. The study confirms the presence of steroids, phenols, glycosides and flavanoids in leaves extract.

Preparation of Extract

Cold Percolation method

The leaves were collected, dried in shade and powdered. About 1g of the powdered leaves was macerated with 1000 ml of double distilled water and tightly covered with aluminium foil and kept for 24 hrs. After 24 hrs, the macerated extract was filtered through Whatmann filter paper. From the extract, the various concentrations were prepared.

Materials and chemicals used

Composition of mild steel

Mild steel specimens (0.026%-S, 0.06%-P, 0.4%-Mn, 0.1%-C and the rest iron) of the dimensions 1x4x0.2 cm were polished to mirror finish and degreased with acetone and used for the weight loss method.

For polarization, cylindrical mild steel rod embedded in Teflon with an exposed area of $1cm^2$ was used. The electrodes were polished with emery papers of 0/0, 2/0, 3/0, and 4/0 grades and degreased with acetone, dried and used.

Acids

1.0 N HCl, was used of G.R. Grade

Weight loss method

Weight loss measurements were performed in 1.0 N HCl with and without the presence of various concentrations was carried out. The inhibition efficiency was calculated by using the formula,

I.E (%) = W_0-W_i/W_0×100

Where,

W_0 = Weight loss in blank

W_i = Weight loss in presence of inhibitor

Potentiodynamic polarization studies

Potentiodynamic polarization studies were performed for mild steel specimens in the presence and the absence of the inhibitors. Polarization measurements were performed to evaluate the corrosion current, corrosion potential and the Tafel slope. The polarization cells contain a three electrode assembly.

Impedance measurements

The instrument used for polarization was also used for AC impedance study. The cell setup was the same as that used for polarization measurements. Cell impedance was measured at various frequencies in ohms. The values of charge transfer resistance, R_t and the double layer capacitance, C_{dl} were calculated.

RESULTS AND DISCUSSION

Weight loss studies

Table-1 gives the inhibition efficiency of different concentrations of *Wattakaka volubilis* leaves extract in 1.0 N HCl. Maximum inhibition efficiency 94.02% is shown by 900 ppm of inhibitor concentration.

Table 1 Inhibition effect on corrosion of mild steel in 1.0 N HCl by *Wattakaka volubilis* leaves extract

[Inhibitor], ppm	Rate of corrosion, g cm^{-2} hr^{-1}	Inhibition Efficiency (%)
Blank	-	-
100	0.000120	20.87
200	0.000110	27.47
300	0.000090	35.79
400	0.000080	40.05
500	0.000068	54.94
600	0.000050	63.73
700	0.000040	73.51
800	0.000020	85.75
900	0.000009	94.02

Adsorption isotherms

Adsorption isotherms are usually used to describe the adsorption process, Anuradha *et al.* (2008). The most frequently used isotherms include: Langmuir, Temkin, Florry-Huggins, and the recently formulated thermodynamic/kinetic model of El-Awady *et al*. The establishment of adsorption isotherms that describe the adsorption of a corrosion inhibitor can provide important clue to the nature of the metal-inhibitor interaction, Ramananada singh *et al*. (2013). Adsorption of the organic molecules occurs as the interaction energy between molecules and metal surface is higher than that between the H_2O molecules and the metal surface.

In order to obtain the adsorption isotherm, the degree of surface coverage (θ) for various concentrations of the inhibitor has been calculated according to its equation. Langmuir isotherm was tested for its fit to the experimental data. Langmuir isotherm is given by

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

Where θ is the degree of surface coverage, C is the concentration of the inhibitor in the bulk solution and K_{ads} is the equilibrium constant of the process of adsorption, Ashassi-Sorkhabi and Asghari 2008.

It is important to know from this part of the study, tables (2-5) and Figs.(2-5) shows the obtained values and graphs were fitted for various isotherms and the best fit was obtained with Langmuir isotherm.

Table 2 Langmuir adsorption isotherm for the inhibition of corrosion of mild steel in 1.0 N HCl using *Wattakaka volubilis* leaves extract

[Inhibitor], ppm	Ɵ/(1-Ɵ)	3+logƟ/(1-Ɵ)	3+log C
Blank	-	-	-
100	0.2637	2.4212	5.0000
200	0.3787	2.5783	5.3010
300	0.5574	2.7462	5.4771
400	0.6681	2.8248	5.6020
500	1.2193	3.0861	5.6989
600	1.7571	3.2448	5.7781
700	2.7750	3.4433	5.8450
800	6.0175	3.7794	5.9030
900	15.7224	4.1965	5.9542

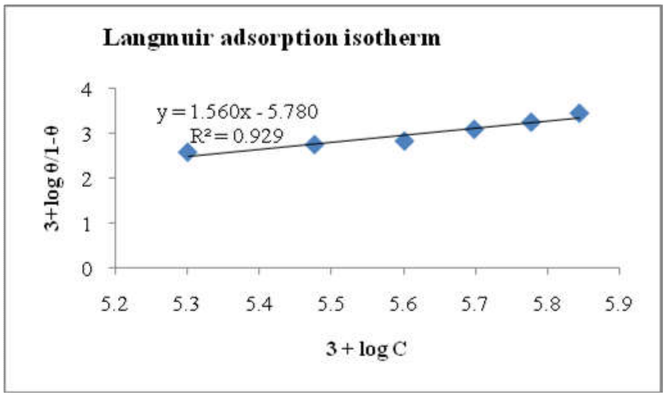

Figure 2 Langmuir adsorption isotherm for the inhibition of corrosion of mild steel in 1.0 N HCl using *Wattakaka volubilis* leaves extract

Table 3 Temkin adsorption isotherm for the inhibition of corrosion of mild steel in 1.0 N HCl using *Wattakaka volubilis* leaves extract

[Inhibitor], ppm	2+log C	Ɵ
100	4.0000	0.2087
200	4.3010	0.2747
300	4.4771	0.3579
400	4.6021	0.4005
500	4.6990	0.5494
600	4.7782	0.6373
700	4.8451	0.7351
800	4.9031	0.8575
900	4.9542	0.9402

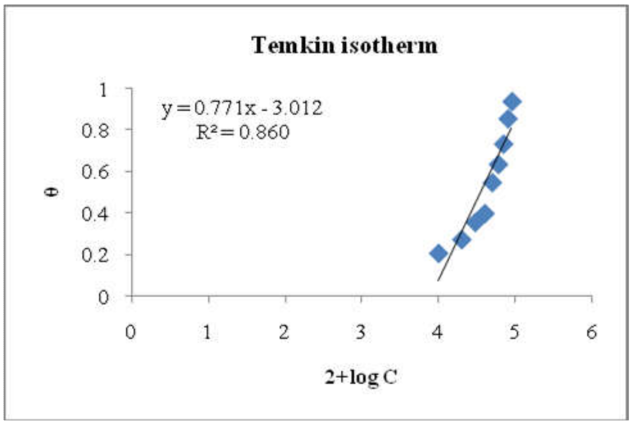

Figure 3 Temkin adsorption isotherm for the inhibition of corrosion of mild steel in 1.0 N HCl using *Wattakaka volubilis* leaves extract

Table 4 Florry-Huggins adsorption isotherm for the inhibition of corrosion of mild steel in 1.0 N HCl using *Wattakaka volubilis* leaves extract

Ɵ	[C], ppm	Ɵ/C	3+logƟ/C	1- Ɵ	3+log(1-Ɵ)
0.2087	100	0.0021	0.3195	0.7913	2.8983
0.2747	200	0.0014	0.1378	0.7253	2.8605
0.3579	300	0.0012	0.0766	0.6421	2.8076
0.4005	400	0.0010	0.0005	0.5995	2.7778
0.5494	500	0.0011	0.0409	0.4506	2.6538
0.6373	600	0.0011	0.0262	0.3627	2.5595
0.7351	700	0.0011	0.0212	0.2649	2.4231
0.8575	800	0.0011	0.0301	0.1425	2.1538
0.9402	900	0.0010	0.0190	0.0598	1.7767

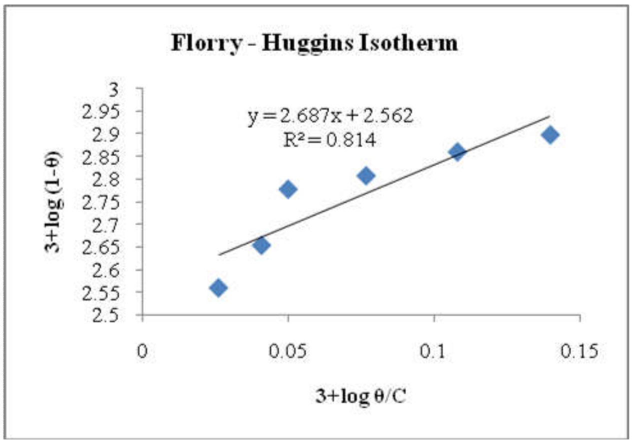

Figure 4 Florry-Huggins adsorption isotherm for the inhibition of corrosion of mild steel in 1.0 N HCl using *Wattakaka volubilis* leaves extract

Table 5 El-awady adsorption isotherm for the inhibition of corrosion of mild steel in 1.0 N HCl using *Wattakaka volubilis* leaves extract

[C], ppm	2+log C	Ɵ	1- Ɵ	Ɵ/(1- Ɵ)	2+log(Ɵ/1-Ɵ)
100	4.000	0.2087	0.7913	0.2637	1.4212
200	4.301	0.2747	0.7253	0.3787	1.5783
300	4.477	0.3579	0.6421	0.5574	1.7462
400	4.602	0.4005	0.5995	0.6681	1.8248
500	4.698	0.5494	0.4506	1.2193	2.0861
600	4.778	0.6373	0.3627	1.7571	2.2448
700	4.845	0.7351	0.2649	2.7750	2.4433
800	4.903	0.8575	0.1425	6.0175	2.7794
900	4.954	0.9402	0.0598	15.7224	3.1965

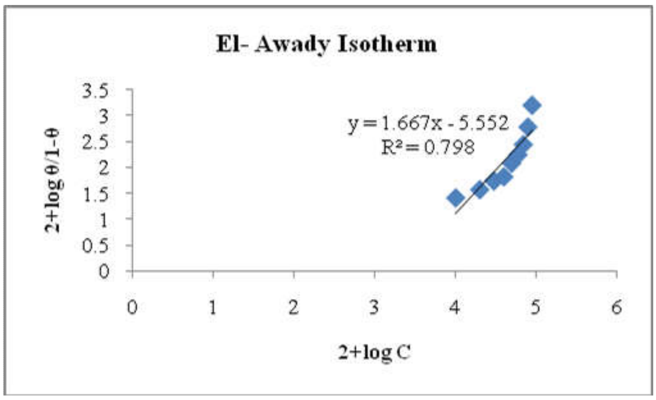

Figure 5 El-awady adsorption isotherm for the inhibition of corrosion of mild steel in 1.0 N HCl using *Wattakaka volubilis* leaves extract

Free energy change

The free energy change of adsorption (ΔG^0_{ads}) of inhibitor on mild steel surfaces related to the constant of the adsorption according to equation, Satapathy *et al*. (2009); Pandian bothi raja and mathur gopalakrishnan sethuraman 2008; Priya *et al*.(2005).

$\Delta G^0_{ads} = -2.303\ RT \log (K_{ads} \times 55.5)$

From the results, showed in table-6 values of ΔG^0_{ads} were found to be negative and were below the threshold value of -40kJ/mol indicating that the adsorption of *Wattakaka volubilis* on mild steel surface is spontaneous and that mechanism of physical adsorption is applicable.

Table 6 Adsorption isotherms parameters for inhibition of corrosion of mild steel in 1.0 N HCl using *Wattakaka volubilis* leaves extract

System	Isotherm	ΔG^0_{ads}	Slope	R^2	a	1/y
Wattakaka volubilis leaves extract	Langmuir Isotherm	-5717.6	1.560	0.929	-	0.641
	Temkin Isotherm	-7365.5	0.771	0.860	-1.542	1.297
	Florry-Huggins Isotherm	-7774.6	2.687	0.814	-	0.372
	El-awady Isothern	-5819.7	1.611	0.798	-	0.620

Potentiodynamic polarization studies

Table-7 and (Figs.5-7) gives the values of potentiodynamic parameters such as corrosion current (I_{corr}), corrosion potential (E_{corr}) and the cathodic Tafel slopes (b_c and b_a) for the different concentrations of green inhibitor understudy. It can be seen that the any one of the components present in the leaves extract adsorbed on metal surface.

AC impedance study

Table-8 and (Figs.8-10) indicates the AC impedance curves and values for inhibitor concentration. The R_t values increases and C_{dl} values decreases with inhibitor concentration. This implies the formation of protective film on the metal surface, Noreen Anthony *et al*.(2004). It can be also seen from table-9 that there is a close agreement between the values of inhibition efficiencies obtained from weight loss measurements, polarization and impedance studies.

Table 7 Corrosion parameters obtained from polarization curves for mild steel in 1.0 N HCl in the presence and absence of inhibitor

System	[Inhibitor], ppm	$I_{corr.}$, mV	$-E_{corr.}$, mV	$b_{a.}$, mV dec^{-1}	$b_{c.}$, mV dec^{-1}	Inhibition Efficiency, (%)
Blank	-	413.79	-484.37	104.8	170.2	-
WATTAKAKA VOLUBILIS Leaves extract	100	332.65	-464.36	113.4	176.8	19.60
	900	40.663	-535.66	173.8	238.7	90.17

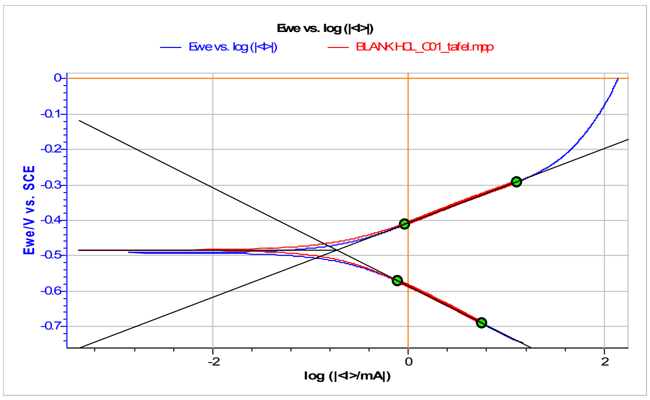

Figure 5 Potentiometric polarization curve for mild steel in 1.0 N HCl

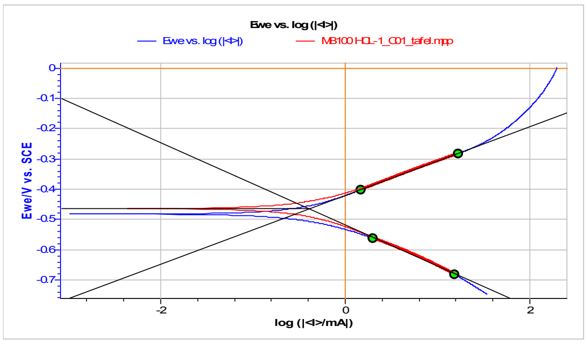

Figure 6 Potentiometric polarization curve for 1.0 N HCl medium in the presence of 100 ppm concentration of *Wattakaka volubilis* leaves extract

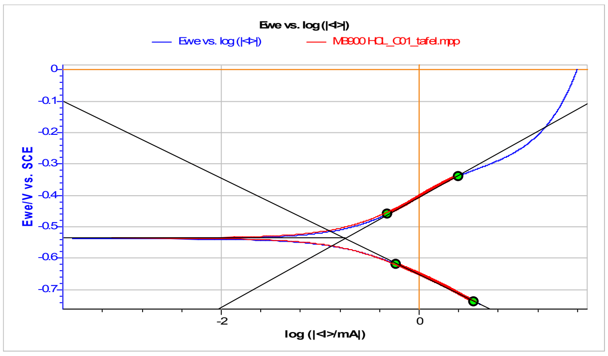

Figure 7 Potentiometric polarization curve for 1.0 N HCl medium in the presence of 900 ppm concentration of *Wattakaka volubilis* leaves extract

Table 8 Corrosion parameters obtained from impedance study for mild steel in 1.0 N HCl in the presence and absence *Wattakaka volubilis* leaves extract

[Inhibitor], ppm	R_t, Ohm,cm^2	C_{dl}, F/cm^2	Inhibition efficiency (%)
Blank	22.24	0.394	-----
100	36.67	0.324	17.76
900	75.00	0.038	90.35

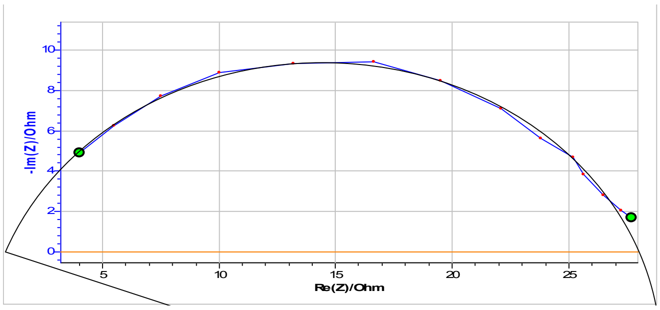

Figure 8 AC impedance diagram for mild steel in 1.0 N HCl

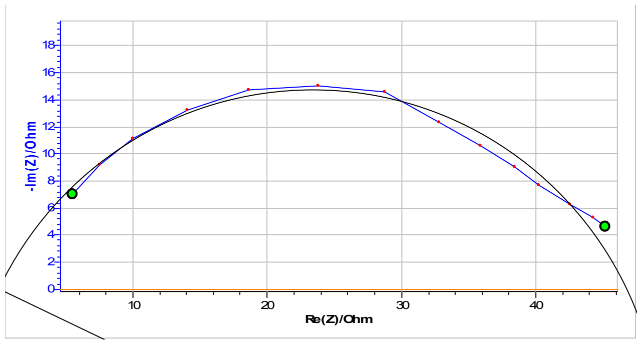

Figure 9 AC impedance diagram for mild steel in 1.0 N HCl medium in the presence of 100 ppm concentration of *Wattakaka volubilis* leaves extract

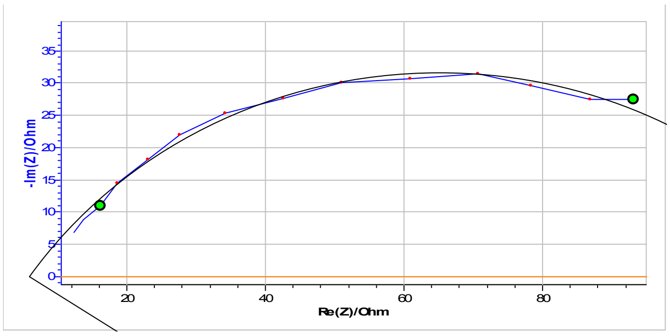

Figure 10 AC impedance diagram for mild steel in 1.0 N HCl medium in the presence of 900 ppm concentration of *Wattakaka volubilis* leaves extract

Table 9 Comparison of inhibition efficiencies measured by weight loss, polarization and impedance studies

[Inhibitor], ppm	Inhibition efficiency (%) Weight loss study	Polarization study	Impedance study
100	20.87	19.60	17.76
900	94.02	90.17	90.35

SUMMARY AND CONCLUSION

The inhibitive influence of *Wattakaka volubilis* leaves extract on the corrosion of mild steel in 1.0 N HCl was studied by weight loss method, polarization and impedance measurements. The inhibition efficiency values determined by these techniques showed close agreement.

- The corrosion decreased with increasing addition of *Wattakaka volubilis* leaves extract probably due to the progressive adsorption of the inhibitor on the metal surface.
- The maximum inhibition efficiency was found to be 94.02% in HCl medium.
- The corrosion inhibition *Wattakaka volubilis* leaves extract is attributed to the adsorption of any of the phyto-chemical components on to the mild steel surface. The adsorption is assumed to arise from the π-bond of the components on the mild steel surface.
- An adsorption isotherm reveals that in HCl, Langmuir isotherm is the best fit for this inhibitor.
- The free energy values for the adsorption processes indicates physisorption of the *Wattakaka volubilis* leaves extract studied on mild steel surface. AC impedance studies reveal that a protective film is formed on the metal surface.
- These results are suggested *Wattakaka volubilis* leaves extract is a best green inhibitor.

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