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Adsorption and corrosion inhibiting behaviour of *Terminalliacattapa* leaf extract on copper steel

Dr. S. Ananthalakshmi[1], A. Anbarasan[2], R. Kavitha[2], S. Kalaivani[2]

1. Research Advisor, 2. Research Scholar, Department of Chemistry,
Urumu Dhanalakshmi College, Tiruchirappalli-19, Tamilnadu, India.

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Abstract

In order to identify new the ecofriendly corrosion inhibitor, the corrosion of copper steel in 0.5 M HCl in the presence and absence of *Terminalliacattapa* leaves extract is estimated. The corrosion rate and inhibitor efficiency of *Terminalliacattapa* leaves extract on the corrosion of copper metal in 0.5M HCl by weight loss measurement were investigated. The effect of temperature on the corrosion behaviour of copper metal in 0.5 M HCl with addition of plant extract and identified the suitable adsorption isotherm for inhibitor molecules. *TerminalliaCattappas* leaf extract acted as a good inhibitor for the corrosion of Copper metal in 0.5 M HCl. Inhibition efficiency values increased with increasing the concentration of plant extract but decreased with temperature. Adsorption of inhibitor molecules on the Copper metal surface was found to obey the Langmuir adsorption isotherm. The corrosion rates were observed to decrease with increase in concentration of plant extract. With increase in temperature from 303 K to 313 K shows a slight decrease in the value of inhibitor efficiency. The degree of surface coverage increases with increase in inhibitor concentration. Thus, the *Terminalliacattappa* leaves extract is an eco-friendly, alternative and commercial use to protect copper metal from the corrosion by the HCl acid.

Introduction

Corrosion can be viewed as a universal problem phenomenon, omnipresent and omnipotent. It is found there everywhere, air, water, soil and in every environment. Corrosion has a huge economic and environmental impact on all facets of national infrastructure; from highways, bridges, buildings, oil and gas lines, chemical processing, water and waste water treatment and virtually on all metallic objects in use. Other than material loss, corrosion interferes with human safety, disrupts industrial operations and poses danger to environment. Awareness to corrosion and adaptation of timely and appropriate control measures hold the key in the abatement of corrosion failures.

Thermodynamic and electrochemical principles play a major role in determining the corrosion behaviour of materials. Thermodynamic principles can indicate the spontaneous direction of a chemical reaction. Thermodynamic principles such as free energy, enthalpy, entropy are used to determine whether the corrosion is theoretically possible or not. [E.C.Potter., (1961),M.Pourbaix.,(1973)].

Electrochemical principles are extensively used to determine the corrosion behaviour of materials. Here, the corrosion reaction can be represented by partial reactions such as metal oxidation and reduction of some reducible species of the environment both occurring simultaneously at equal rates, at the mixed potential of the reaction. [C.Wagner et al.,(1938)] . Corrosion reaction mainly occurs at the metal-environment interface. The electrochemical nature of corrosion can be illustrated by the attack of iron in hydrochloric acid. When the iron is dipped in acid a vigorous action occurs as a result hydrogen gas is evolved and iron gets dissolved.Hence the reaction is

$$Fe + 2H^{+} \leftrightarrows Fe^{2+} + H_2 \rightarrow (1)$$

The above reaction (1) can be divided into two partial reactions,

Oxidation (anodic) reaction: $Fe \longrightarrow Fe^{2+} + 2e^{-}$ →(2)

Reduction (cathodic) reaction: $2H^{+} + 2e^{-}$ $H_2 \longrightarrow$ →(3)

Inhibitors are chemicals that react with a metallic surface, or the environment this surface is exposed to, giving the surface a certain level of protection. Inhibitors often work by adsorbing themselves on the metallic surface by forming a film. Inhibitors are normally distributed from a solution or dispersion. By definition, a corrosion inhibitor is a chemical substance that, when added in small concentration to an environment, effectively decreases the corrosion rate. The efficiency of that inhibitor is thus expressed by a measure of this improvement.

$$\text{Inhibitor Efficiency (\%)} = \frac{(CR_{uninhibited} - CR_{inhibited})}{CR_{uninhibited}} \times 100 \rightarrow (4)$$

Where, $CR_{uninhibited}$ = corrosion rate of the uninhibited system

$CR_{inhibited}$ = corrosion rate of the inhibited system

In general, the efficiency of an inhibitor increases with an increase in inhibitor concentration. Inhibitors are substances or mixtures that in low concentration and in aggressive environment inhibit, prevent or minimize the corrosion. [I.B. Obotet al.,(2009)][V. Gentil.,(2003)] [Hong Ju et al.,(2008)] [L. V. Ramanathan.,(1988)]

Terminaliacatappa

The trees are monoecious, with distinct male and female flowers on the same tree. The fruit is a drupe 5–7 cm (2.0–2.8 in) long and 3–5.5 cm (1.2–2.2 in) broad, green at first, then yellow and finally red when ripe, containing a single seed. During the dry season, the leaves turn into autumn colours of red, copper, gold. The tree usually sheds all its leaves twice a year in January-February and July-August. The tree first drops its leaves when it reaches 3-4 years old.

Figure.1. Flower *Terminaliacatappa*

Metals are affected by different forms of corrosion in various environmental conditions. The exposures can be most severe, but in many cases the corrosion may be controlled by means of inhibitors. Use of inhibitors in these processes to prevent metal dissolution is very common. Most of the well-known acid inhibitors are organic compounds containing nitrogen, sulfur, oxygen, heterocyclic compounds with a polar functional group and conjugated double bonds. These compounds are adsorbed on the metallic surface and block the active corrosion sites.

Most of the synthetic chemicals are costly, toxic to both human being and the environment. To solve the above mentioned defects, it is necessary to develop cheap, non-toxic and environmentally friendly natural products as corrosion inhibitors. Naturally occurring compounds from bio-materials can be used as inhibitors in acid solution. In recent years, plant extracts have become the focus of corrosion inhibitor research due to their low toxicity, easy availability and economical preparation. These natural organic compounds are either synthesized or extracted from aromatic herbs, spices and medicinal plants. Plant extracts are an incredibly rich source of naturally synthesized chemical compounds that can be extracted by simple procedures with low cost and are easily biodegradable in nature.

Experimental methods

i) Electrode Preparation

A copper steel specimen of Size was used. The plates were washed, dried and polished successively using emery sheets of extra coarse, coarse, medium, find and extra fine grades to removes adhering impurities and finally degreased with acetone and dried with a drier. The plates were then kept in desiccators to avoid the absorption of moisture by the plates.

ii) Experimental Procedure

The initial weight of the polished plate was taken. The 0.5M HCl were taken in a 100 ml beaker and the specimens were in dipped into the solution using glass hooks. Care should be taken to ensure the complete immersion of the specimen. After a period of one hour, the specimens were removed, washed with water, dried, and weighted to an accuracy of four decimals. From the initial and final masses of the specimen (before and after immersion in the solution) the loss in weight was calculated. The experiment was repeated for various inhibitor (plant extract) concentrations. To know the effect of temperature the above procedure was carried out at different temperature ranges i.e 303, 313 K using a thermostat, for 0.5M HCl with different inhibitor concentration.

Results and discussion

The inhibition efficiency of corrosion on copper metal using 0.5 M HCl with five different concentration of inhibitor have been studied by weight loss methods. The nature of adsorption has also been obtained by studying the behavior of inhibitor at various temperature (303 K & 313 K) using weight loss techniques. The I.E in the presence and absence of inhibitor has also been studied using weight loss measurements. The I.E, corrosion rate, the nature of anchoring sites and the adsorption characteristics have been determined from the results obtained.

Weight loss studies

Weight loss method has been carried out at various concentrations (1 ppm to 5 ppm) of the inhibitor in 0.5 M HCl. In this study the parameters like corrosion rate (mpy) and surface coverage (θ) have been calculated. The effect of temperature has studied by varying the temperature from 303 K and 313 K.(Table.1)

Effect of concentration of inhibitor on inhibition efficiency, weight loss and corrosion rate

The study revealed that the plant extract (*Terminaliacatappa*) inhibited the corrosion of copper metal even at low concentration. The inhibition efficiency has found to be increase with increase in inhibitor concentration. It has been observed that the plant extract inhibit the corrosion of copper metal at all concentration used in our study.

Inhibitor efficiency has been determined by using the following relationship

$$\text{I.E (\%)} = \frac{(W_o - W_i)}{W_i} \times 100 \qquad \rightarrow (5)$$

Where Wo is the weight loss without inhibitor and W_i is the weight loss with inhibitor.

The effect of concentration of inhibitor on weight loss measurements has been conducted for uninhibited and inhibited systems in 0.5M HCl for a period of 1 hour. A Plot of weight loss Vs inhibitor concentration revels that the metal loss has found progressively decreased with increasing inhibitor concentration.

Corrosion rate has been determined from the following relationship

$$\text{Corrosion rate} = \frac{534 \times W}{D \times A \times T} \qquad \rightarrow (6)$$

Where,

W= Weight loss in mg
D = Density in g/cc

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A = Area of exposure in Sq. inches

T= Time in hours

The corrosion rate in 0.5M HCl for various concentrations of the inhibitors was determined after 1 and 3 hours of immersion. The corrosion rate expressed in mpy and it is decreased within increase in inhibitor concentration as evident from Tables(2,3),Figures (1,2)

Effect of concentration of inhibitor on surface area

The surface coverage (θ) for different inhibitor concentrations were calculated. The observations of a plot C/ θ Vs C gives a straight line confirming that the inhibitor obeyed Langmuir adsorption isotherm. This result supports conclusion that maximum inhibition correspond to the formation of an adsorbed layer of the inhibitor on the active sites of the metal surface.

$$\text{The surface coverage } (\theta) = \frac{(W_b - W_i)}{W_b} \times 100 \qquad \rightarrow (7)$$

Where, W_b and W_i are the weight loss per unit time with and without inhibitor respectively. A graph a was drawn between C/ θ Vs C or C/ θ Vs Log C to know whether the adsorption of inhibitor follows Langmuir isotherm to obtain linear relationship.

Influence of temperature

The effect of temperature on corrosion rate and inhibition efficiency (I.E) using weight loss methods were carried out at two different temperatures (303K, 313K) in the absence and presence of inhibitor at selected concentration of 0.5M HCl and the results are tabulated. (Table 2,3),Figures(3,4)

It is evident from the results obtained (Table 2,3) that in 0.5M HCl the dissolution of metal increase with rise in temperature both in absence and presence of inhibitor and efficiency of inhibitor decrease with increase in temperature indicating weak adsorption. The decrease in I.E with temperature indicates, the fact that the inhibitor film formed on the metal surface is less protective in nature at higher temperature.

Table 1 – Weight loss and Corrosion rate data for copper metal in HCl

Concentration of HCl (M)	Weight of the metal		Weight loss (g)	Corrosion rate (mmy^{-1})
	Initial (g)	Final (g)		
0.5	7.032	7.010	0.022	9.433
0.4	7.002	6.984	0.018	7.718
0.3	6.978	6.962	0.016	6.860
0.2	6.951	6.941	0.010	4.288
0.1	6.935	6.927	0.008	3.430

Corrosion rate (mmy^{-1}) = 87.6 W / DAT
W = Weight loss in mg
D = Density in g cm^{-3}
A = Area of sample in cm^2
T = Time of exposure of the metal sample in hours.

Table 2 – Inhibition efficiency of different concentration of *TerminaliaCatappa* leaf extract for corrosion of copper metal in 0.5M HCl solution at 303K

Concentration of HCl (M)	Concentration of inhibitor (ppm)	Weight loss (g)	Corrosion rate (mmy^{-1})	Surface coverage (θ)	Inhibitor efficiency IE(%)
0.5	-	0.022	9.433	-	-
0.5	1	0.017	7.289	0.2272	22.72
0.5	2	0.013	5.574	0.4090	40.90
0.5	3	0.010	4.288	0.5454	54.54
0.5	4	0.008	3.430	0.6363	63.63
0.5	5	0.006	2.572	0.7272	72.72

Table 3 – Inhibition efficiency of different concentration of *TerminaliaCatappa* leaf extract for corrosion of copper metal in0.5M HCl solution at 313K

Concentration of HCl (M)	Concentration of inhibitor (ppm)	Weight loss (g)	Corrosion rate (mmy^{-1})	Surface coverage (θ)	Inhibitor efficiency IE(%)
0.5	-	0.046	19.725	-	-
0.5	1	0.039	16.723	0.1521	15.21
0.5	2	0.033	14.150	0.2826	28.26
0.5	3	0.027	11.577	0.4130	41.30
0.5	4	0.022	9.433	0.5217	52.17
0.5	5	0.017	7.289	0.6304	63.04

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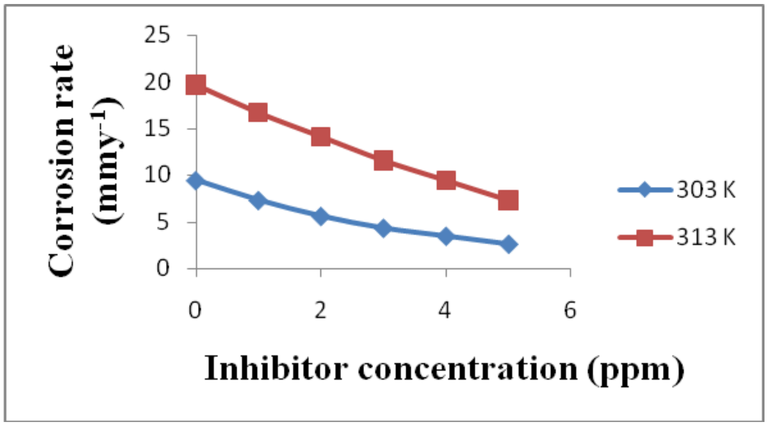

Figure.1 – Effect of concentration of *TerminaliaCatappa* leaf extract on corrosion rate of copper metal in 0.1 M HCl at different temperature

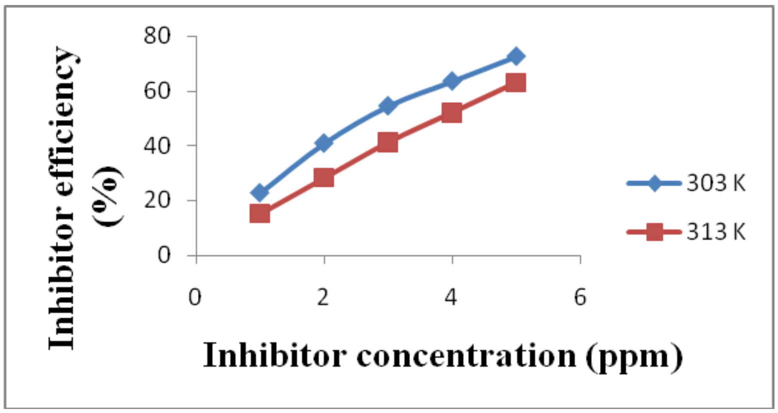

Figure.2 – Effect of concentration of *TerminaliaCatappa* leaf extract on inhibitor efficiency of copper metal in 0.1 M HCl at different temperature

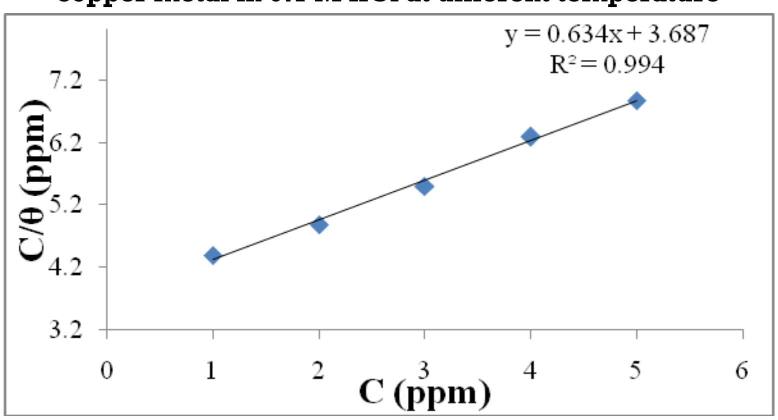

Figure.3 – Langmuir adsorption isotherm plot for copper metal in 0.5 M HCl at 303 K in the presence of various concentration of *TerminaliaCatappa leaf* extract

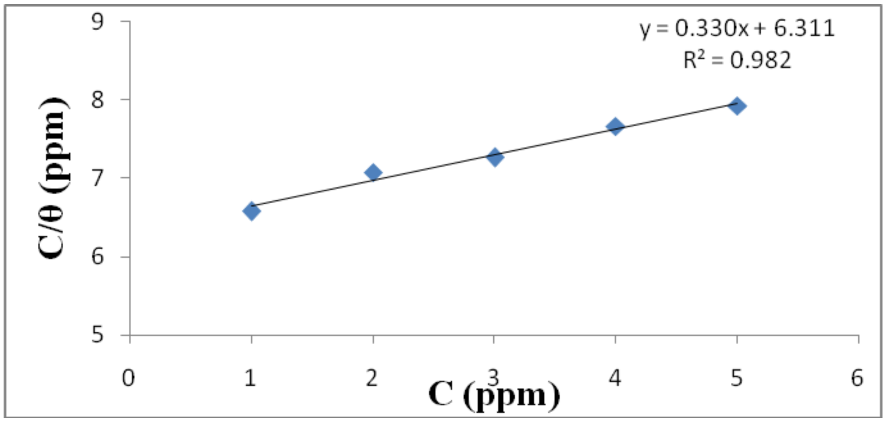

Figure.4 – Langmuir adsorption isotherm plot for copper metal in 0.5 M HCl at 313 K in the presence of various concentration of *TerminaliaCatappa leaf* extract

Conclusion

Extracts of plant materials are less expensive and environmentally friendly natuure. Extracts of plant materials contain many active principles for its activity. They contain polar atoms such as S, N, O, P etc. Because of this nature, the lone pair of electrons present on these atoms is pumped on to the metal surface; loss of electrons from the metal surface can be avoided. Thus, corrosion inhibition takes place. Because of adsorption of inhibitor molecules on metal surface, protective film is formed and hence corrosion is controlled.

TerminalliaCattappas leaf extract acted as a good inhibitor for the corrosion of Copper metal in 0.5 M HCl. Inhibition efficiency values increased with increasing the concentration of plant extract but decreased with temperature. Adsorption of inhibitor molecules on the Copper metal surface was found to obey the Langmuir adsorption isotherm.

The following conclusions can be drawn from the present study

- Weight loss measurement results shows that the plant extract act as a excellent corrosion inhibitor.
- The corrosion rates were observed to decrease with increase in concentration of plant extract.
- With increase in temperature from 303 K to 313 K shows a slight decrease in the value of inhibitor efficiency which is due to the removal of loosely adsorbed molecules by mechanical vibration of thermal energy.
- The degree of surface coverage increases with increase in inhibitor concentration. This was attributed to more adsorption of inhibitor molecules on to the surface of the copper metal.

- Adsorption isotherms provide a basic information on the interaction between the inhibitor and mild steel surface. The experimental data shows that the adsorption of the studied inhibitor on copper metal obeyed Langmuir adsorption isotherm.
- The evaluation stated that the *Terminalliacattappa* leaf extract is an eco-friendly, alternative and commercial use to protect copper metal from the corrosion by the HCl acid.

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