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Full Length Research Paper

# Uvaria chamea root as green corrosion inhibitor for mild steel in acidic solution

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The corrosion inhibition of mild steel is the subject of tremendous technological importance due to the increased industrial applications of these materials. This study reports the corrosion inhibition of mild steel in acidic solution of 0.5 M HCl using root extract of *Uvaria chamea* by way of gravimetric measurements. It was shown that the presence of *U. chamea* root extract inhibited the corrosion of mild steel in the test solution and the inhibition efficiency depended on the concentration of the plant extract as well as on the time of exposure of the mild steel samples in HCl solutions containing the extract. The experimental data complied to the Langmuir and Temkin adsorption isotherms. From the results obtained authenticates that the root extracts of *U. chamea* favours both cathodic and anodic inhibitor. The adsorption mechanism is both chemisorptions and physisorption.

Key words: Corrosion inhibition, mild steel, adsorption mechanism, Uvaria chamea, Langmuir and Temkin isotherms.

# INTRODUCTION

The use of inhibitors is one of the most practical methods for protecting metals against corrosion, especially in acidic media (Trabanelli, 1991). Acid solutions are widely used in industry: some of the important fields of application are acid pickling of steel, chemical cleaning and processing, ore production and oil well acidizing. As ordinary acids, HCI and  $H_2SO_4$  are usually used as industrial acids, cleaning and pickling acids. Due to the general aggression of acid solutions, inhibitors are commonly used to retard the corrosive attack on metallic materials. During past decades, some commercial inhibitors have been synthesized and used successfully to inhibit corrosion of steel in acidic media. However, the major problem associated with most of these inhibitors is that they are not eco-friendly but toxic and expensive. Therefore, the study of new non-toxic corrosion inhibitors is essential to overcome this problem. The research in the field of eco-friendly corrosion inhibitors has been addressed toward the goal of using cheap, effective compounds at low or "zero" environmental impact (Yadav and Sharma, 2011).

Plant extracts are low-cost and biodegradable, and so the study of plant extracts as corrosion inhibitors is an important scientific research field due to both economic and environmental benefits. It has been found that certain organic substances containing polar functions with nitrogen, sulphur and/or oxygen atoms in the conjugated system have been reported to exhibit good inhibiting

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properties of steel in acidic and alkaline environments (Popova et al., 2003; Oguzie, 2008a, b; Ebenso et al., 2009; Ulaeto, 2012; Atanda, 2012). The results of parallel studies of the inhibiting effects of organic compounds suggest that the inhibitory behavior of the organic compounds subsists in some chemical species or molecules in the inhibiting substances forming a protective layer between the metal surface and the corrodents. The adsorbate layer formed isolates the metal surface from the corrodents thereby reducing the corrosion rate of the metal surface. It has been recognized that the use of organic inhibitors, particularly the naturally occurring organic inhibitors of plant origin, are viable and highly beneficial since they are essentially non-toxic, environmentally benign, readily available, renewable and inexpensive (Oguzie, 2008a, b; Ebenso et al., 2004, 2009; Martinez, 2002; Abiola et al., 2007; Chauhan and Gunasekaran, 2007; Mejeha et al., 2010; Behpour et al., 2012: Deng and Li, 2012: Educk et al., 2012; Garai et al., 2012; Gerengi and Sahin; 2012; Li et al., 2012a; Li et al., 2012b; Soltani et al., 2012). Through these studies, it is agreed that the inhibition performance of plant extracts is normally ascribed to the presence, in their composition, of complex organic species such as tannins, alkaloids and nitrogen bases, carbohydrates, amino acids and proteins as well as hydrolysis products. These organic compounds contain polar functions with N, S, O atoms as well as conjugated double bonds or aromatic rings in their molecular structures, which are the major adsorption centre. In the present work, Uvaria chamea root extract is chosen to be the corrosion inhibitor.

U. chamea is a climbing plant predominantly found in the tropical rain forest of West Africa (Okwu and Iroabuchi, 2009; Irvin, 1961). The root extracts have been known to have various medicinal and therapeutic properties, and nutritional values. Phytochemical studies on the plants revealed the presence of bioactive components comprising flavonoids (0.70 to 5.70 mg. 100  $g^{-1}$ ), alkaloids (0.81 to 5.40 mg. 100  $g^{-1}$ ), tannins (0.40 to 3.60 mg. 100  $g^{-1}$ ), saponins (0.38 to 2.10 mg. 100  $g^{-1}$ ) and phenols (0.08 to 0.10 mg. 100  $g^{-1}$ ) (Soltani et al., 2012). They are used in Nigeria as medicinal plants in phytomedicine to cure diseases and heal injuries. These plants have various effects on living systems. They are sedative, analgesic, cardio-protective, anti-inflammatory, oxytocic, antispasmodic and immune modulators (Okwu, 2007).

#### MATERIALS AND METHODS

#### **Material preparation**

The sheets of mild steel used for this study were obtained from Federal University of Petroleum Resources, Effurun- Warri, Nigeria. Each sheet was 1.32 mm in thickness and were mechanically pressed cut into  $2 \times 2$  cm coupons. These coupons were used as cut and polished to remove the corroded surface. However, they

were degreased in ethanol, dried in acetone and stored in moisture free desiccators before their use in corrosion studies.

#### Preparation of the root extracts of *U. chamea*

The procedure for the preparation of the root extracts is similar to that reported recently by Okafor et al. (2008). *U. chamea* roots were collected from Umuahia, Abia State, Nigeria. They were dried in an N53C-Genlab Laboratory oven at 50°C, and ground to powder form. Ten gram of the powder was digested in 1L of 0.5 M HCl solution. The resultant solution was kept for 24 h, filtered and stored. From the stock solution, the root extracts test solutions were prepared at concentration range of 0.1 to 0.4 g/L using excess acid as solvent at room temperature and 60°C using water bath.

#### Gravimetric experiment

The cleaned and dried specimens were weighed before immersion into the respective test solutions of 0.5 M HCl using JA 1003A electronic weighing balance with the accuracy of  $\pm 0.005$ . Tests were conducted with different concentrations of inhibitor. At the end of the tests, the specimens were carefully washed in absolute ethanol having used nitric acid to quench further corrosion from taking place, and then reweighed. Triplicate experiments were performed in each case and the mean values reported.

# **RESULTS AND DISCUSSION**

# Fourier transform infrared spectroscopy (FTIR)

The Fourier Transform Infrared Spectroscopy (FTIR), was employed (Figure 1) to determine the functional groups present in the organic compound of the inhibitor, U. chamea. U. chamea was subjected to IR radiation, the energy absorbed results in molecular vibrations in bonds between atoms in the molecule. The bond between two atoms are likened to springs that have stretching and bending properties, while the atoms are regarded as tiny masses attached to both ends of the spring. It is also important to note that for IR absorption to take place, the bond vibration must result in a change in the dipole moment of the bond. If the stretching or bending vibration of a bond results in a large change in its dipole moment, the bond will show strong absorption bands in the IR spectrum. If the change in dipole moment is not significant, a weak absorption band will result.

The absorption of IR radiation results in the elevation of the molecule to a higher vibrational energy level. 3422 cm<sup>-1</sup> signifies H - C - H asymmetric and symmetric stretch which belong to the functional group alkenes (alcohol and phenols) while 2931 cm<sup>-1</sup> is the C – H stretch of alkanes. The peak value of 1640 cm<sup>-1</sup> is the C = O stretch which belongs to the functional group of aldehydes and saturated aliphatic and the vibration is in conjugation with oxygen in the R portion shifts the absorption to higher frequencies. 1347 cm<sup>-1</sup> is the C – N stretch of aliphatic amines. From the interpretation of the IR result, the absorption rate of *U. chamea* as an inhibitor is quite high with the presence of the C = O functional group.

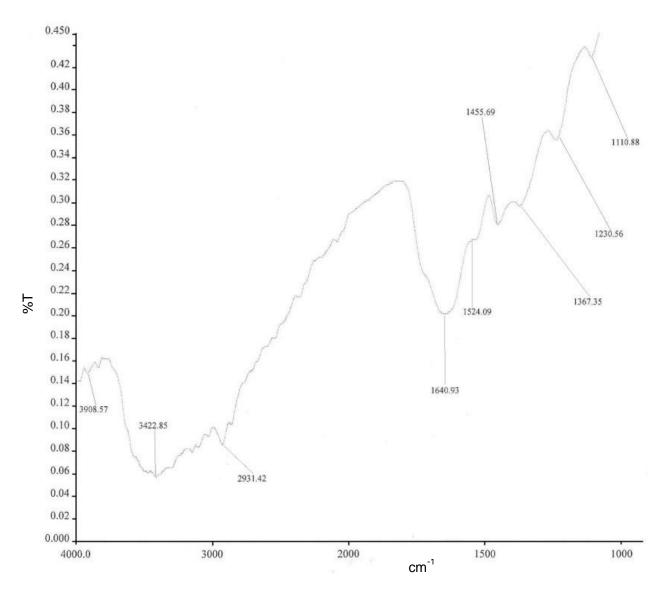


Figure 1. Fourier Transform Infrared Spectroscopy U. chamea root extract.

#### Gravimetric technique and corrosion rates

The corrosion rate values for the mild steel in 0.5 M HCl solutions in the absence and presence of *U. chamea* root extract were determined at room temperature. Figure 2 illustrates the variation of the corrosion rates of the mild steel in 0.5 M HCl with inhibitor concentration for an exposure time of 7 h. The figure clearly shows that *U. chamea* root extract retards the corrosion rate of the mild steel in the test solutions. Moreover, it can be seen from Figure 2 that the corrosion rate decreases with increase in the concentration of the inhibitor. The equation for corrosion rate is given by

$$C = \frac{K\Delta W}{\rho At} \tag{1}$$

where C is the corrosion rate,  $\Delta W$  is the weight loss in mg;  $\rho$  is the density of the Al (g cm<sup>3</sup>); A is the exposed area of the coupon (in<sup>2</sup>; 1 in<sup>2</sup> = 6.5146 cm<sup>2</sup>); t is the immersion time (h); K is the rate constant (534 mpy; mils per year; 1 mil = 10<sup>-3</sup> in).

The inhibition efficiency of the *U. chamea* root extract on the corrosion of the mild steel in 0.5 M HCl containing different concentrations of the root extract was computed by using the relation (Oguzie, 2007; Oguzie et al., 2007).

$$I\% = 1 - \frac{\rho_{inh}}{\rho_{blank}} \times 100 \tag{2}$$

where I% represents the inhibition efficiency expressed in percentage,  $\rho_{\text{inh}}$  is the corrosion rate in the presence of the inhibitor while  $\rho_{\text{blank}}$  is the corrosion rate in the

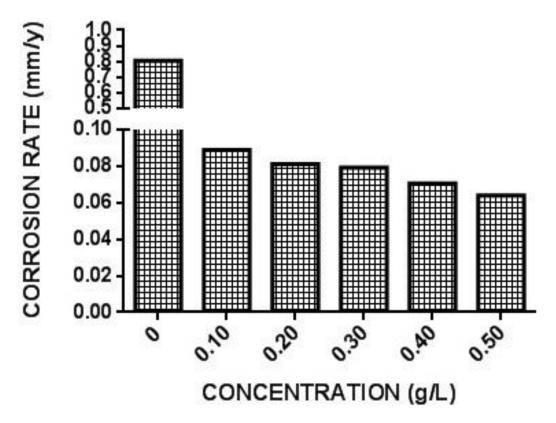


Figure 2. Variation of corrosion rate of mild steel in 0.5 M HCl with different concentrations of U. chamea extract.

absence of inhibitor. They show high levels of inhibition efficiency with optimum value of 92% at 0.5 g/L 0.5 M HCl for concentration of *U. chamea* extract was obtained as shown in Figure 3. The results show that *U. chamea* root extract are good.

# Inhibition efficiency

Clearly, the inhibition efficiency of *U. chamea* root extract increases with exposure time for the inhibitor concentration considered. As shown in Figure 3, the inhibition efficiency increases with increase in inhibitor concentration, tending to saturate at higher values of inhibitor concentration. This indicates that the effectiveness of the *U. chamea* root extract in retarding the corrosion rate of mild steel in the test solutions does not improve indefinitely with increase in inhibitor concentration.

The result that the inhibition efficiency of *U. chamea* root extracts increase with increase in inhibitor concentration suggests that some of the molecules of the inhibitor are adsorbed on the metal surface thereby protecting the "covered" surface from further corrodent attack. Increasing the inhibitor concentration increases the degree of surface coverage,  $\theta$ , of the metal surface

defined as:

$$\theta = 1 - \frac{\rho_{inh}}{\rho_{blank}} \tag{3}$$

The *U. chamea* root extract consists of a mixture of complex organic components including flavonoids, several phenolic compounds ( $\beta$ -carotene – linoleic acids) and some other organic compounds (Okwu and Iroabuchi, 2009). The plant extract also contains some nutritional minerals including potassium, magnesium, copper and zinc (Trabanelli, 1991; Popova and Hincha, 2004; Popova et al., 2003; Popova, 2007).

It is interesting to note that a number of the above named organic compounds exhibit antioxidant activity. For example, nasunin (a flavonoid), g-carotene – linoleic acids (phenolic compounds) are antioxidants. Results of earlier studies suggest that tannins and some antioxidant compounds in plant extracts contribute to inhibition of steel corrosion in some acidic medium (Popova, 2007; Ebenso et al., 2004; Raghavan, 1989). In fact, plant/leaf extracts which have proven corrosion inhibiting abilities in corrosive media are known to contain one or more of the following organic substances. namely tannins triterpenoids, flavonoids, amino acids, alkaloids, saponins,

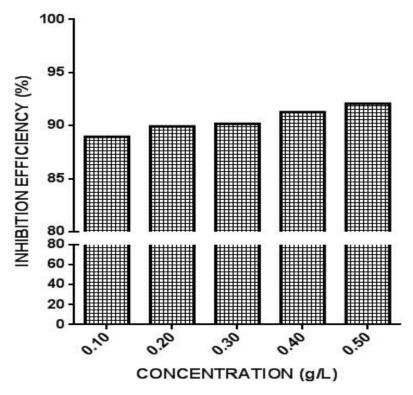


Figure 3. Variation of inhibition efficiency with concentration of *U. chamea* root extract for mild steel in 0.5 M HCl.

phenols, glycosides, essential oils, carotenoids, ßcarotene, ascorbic acid, crude proteins among others (Popova and Hincha, 2004; Popova et al., 2003; Popova, 2007; Noor, 2009). Some research workers Raja and Sethuraman (2008) and Rajendran et al. (2000) have demonstrated that amino acids are indeed effective corrosion inhibitors. It is therefore reasonable to expect that the presence of amino acids, antioxidant organic compounds (nasunin - a flavonoid, caffeic and chlorogenic acids -phenolic compounds), crude proteins or indeed any other of the complex organic substances in the Uvaria chamea root extract could in combination with the other constituents be responsible for the inhibition of mild steel corrosion in 0.5 M HCl. Moreover, the presence of some metallic ions particularly Mg<sup>2+</sup> may enhance the corrosion inhibiting action of the extract. This is because it has been observed that some inorganic ions particularly  $Ca^{2+}$ ,  $Mg^{2+}$  and  $Zn^{2+}$  ions synergistically increase the inhibition efficiency of organic substances (Trabanelli, 1991, Sajise et al., 1974; Sharma et al., 2008). Nevertheless, it is not possible at this point to identify the particular constituent or group of constituents of the plant extract that are adsorbed onto the metal surface. In any case, the adsorbate molecules on the metal surface constitute a barrier to charge and mass transfer between the metal and the corrodent, thereby protecting the metal surface from corrodent attack. The larger the degree of surface coverage resulting from enhanced adsorption of molecules of the plant extract, the greater the protection to corrosion offered by the inhibitor (Oguzie et al., 2007).

## Adsorption considerations and adsorption isotherms

In the situation where it is suspected that the inhibition of metal corrosion occurred as a result of the adsorption of molecules of plant extracts onto the metal surface, it is instructive to investigate the possible adsorption mode by testing the experimental data obtained with several adsorption isotherms.

Such an exercise will greatly elucidate one's understanding of the corrosion inhibition mechanism. The generalized expression for several adsorption isotherms usually tested is of the form (Okafor et al., 2008; Sudheesh, 1999; Trease and Evans, 2002).

$$f(\theta, x) \exp(-\alpha \theta) = kC \tag{4}$$

where f ( $\theta$ , *x*) is the configuration factor whose functional form depends on the physical model adopted and assumptions made in deriving the isotherm,  $\theta$  is the degree of surface coverage, *x* is known as the size ratio which gives the number of water molecule replaced by the inhibitor molecule,  $\alpha$  is a molecular interaction parameter whose value depends on the type of molecular

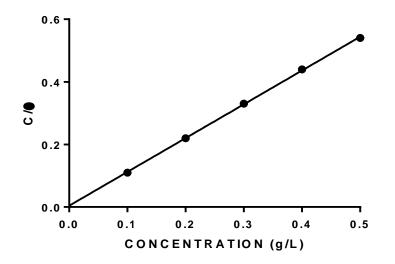


Figure 4. Langmuir adsorption isotherm for inhibition of mild steel in 0.5 M HCl by *U. chamea* root extract.

interactions in the adsorption layer and the degree of homogeneity of the surface, C is the inhibitor concentration while k is the adsorption equilibrium constant which is temperature dependent according to the relation (Okafor et al., 2008; Trease and Evans, 2002).

$$k = \frac{1}{55.5} \exp\left(-\frac{\Delta G_{ads}^{\circ}}{RT}\right)$$
(5)

where  $\Delta G_{ads}$  is the standard free energy of adsorption, R, is the molar gas constant and T is absolute temperature. Several adsorption isotherms were tested for fit with the experimental data. These include the Langmuir, Frumkin, Temkin, Freundlich and the Flory-Huggins isotherms. Incidentally, the Langmuir isotherm gave the best fit with the experimental data. The Langmuir isotherm equation is of the form (Oguzie et al., 2007; Raghavan, 1989; Sharma et al., 2008; Sudheesh, 1999; Trease and Evans, 2002; Umoren et al. 2006; Sofowora, 1993; Sudheesh et al., 1997).

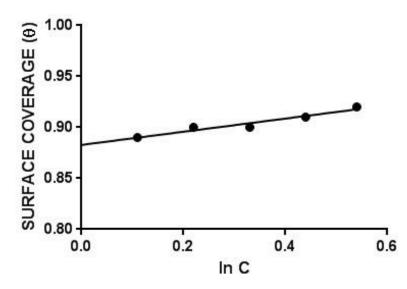
$$\frac{C}{\theta} = \frac{1}{k} + C \tag{6}$$

From a plot of C/ $\theta$  against C, a straight line graph was obtained with a slope of 1.080 and an intercept of 0.004 was obtained. The coefficient of correlation R<sup>2</sup>, gave the degree of fit between the experimental data and the isotherm equation. The value obtained was found to be 0.9997 which indicates a very good fit between the Langmuir isotherm and the experimental data. Figure 4 shows the Langmuir isotherm plot for the inhibition of the corrosion of mild steel in 0.5 M HCl by *U. chamea* root extract. From the plot, one can infer that physisorptions occurred.

The adsorption of uncharged molecules on a heterogeneous surface is appropriately described by the Temkin isotherm [ $\theta$  = (1/f) In kadsC], where  $\theta$  is a linear function of In C (Fuchs-Godec and Zerjav, 2009). This isotherm contains a factor (f) that clearly takes in account adsorbent-adsorbates interactions and considers the fact that the adsorption heat of all molecules in the layer decreases with coverage due to the adsorbate-adsorbent interaction. This makes the isotherm quite suitable for systems where chemical interaction (chemisorptions) of inhibiting species with the metal surface is more pronounced. Temkin adsorption isotherm assumes a uniform distribution of adsorption energy, which decreases with the increase of the value of surface coverage,  $\theta$ . The Temkin adsorption isotherm, which favours chemisorption was investigated, and the data shown in Figure 5. Figure 5 shows straight line chart with high correlation coefficient. The values obtained agree with literature survey which suggests chemisoprtion (Umoren et al., 2008; Ebenso et al., 2009; Obot et al., 2010). The values of  $\Delta G_{ads}$  obtained (Table 1) also confirm chemisorptions.

### Conclusion

Adsorption characteristics of the plant extracts were approximated by the Langmuir and Temkin isotherms. The Langmuir and Temkin isotherms were obeyed which authenticates the physisorbed and chemisorbed adsorption mechanism. The thermodynamic parameter applied also confirms this. From the results, it is obvious that *U. chamea* root extract is a mixed inhibitor of both cathodic and anodic. Therefore, *U. chamea* root extract is a good corrosion inhibitor of mild steel.



**Figure 5.** Temkin adsorption isotherm for inhibition of mild steel in 0.5M HCl by *U. chamea* root extract.

Table 1. Adsorption parameters of U. chamea effect on mild steel in acidic solution.

Isotherms	Slope	Intercept	К	R <sup>2</sup>	∆G <sub>ads</sub> (kJmol <sup>-1</sup> )
Langmuir	1.0800	0.0040	2.5000×10 <sup>2</sup>	0.9997	-24,027
Temkin	0.0167	0.9264	1.2349×10 <sup>24</sup>	0.8615	-149,862

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