

Inhibition of Mild Steel Corrosion by *Aspilia africana* in Acidic Solution

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Abstract This paper is a study of the inhibitive effect of *Aspilia africana* leaves extract on mild steel corrosion in 1.0 M hydrochloric acid (HCl) at room temperature and at 60°C. The leaves extract had shown good inhibition efficiency on the corrosion of mild steel with optimum values of 88.1% at room temperature and 91% at 60°C. The Langmuir and Temkin isotherms were used to analyse the adsorption mechanism of the inhibitor-metal interaction. The Gibb's free energy of adsorption showed a spontaneous and mixed adsorption-physisorption and chemisorption. The deductions from the apparent activation energy and heat of adsorption calculations also confirmed a mixed adsorption mechanism.

Keywords Adsorption, Chemisorptions, Effects of temperature, Acidic medium, Corrosion

1. Introduction

The corrosion of metals is a serious problem in many industries, installations and civil services such as water and sewage supplies. One of the most useful and practical methods that is used to control and protect metals against corrosion is the use of inhibitors, especially in acidic media. Not only the costs due to corrosion increase rapidly, but also the efficiency of the plants and the quality of the products is also reduced. Most inhibitors are organic compounds containing polar groups strongly adsorbed on the metal surface [1, 2, 3]. These inhibitors, which include the organic nitrogen (N), phosphorus (P), sulphur (S), and hydroxy (OH) groups, are known to be similar to catalytic poisons, as they decrease the reaction rate at the metal/solution interface without being involved in the reaction considered. It is generally accepted that most organic inhibitors act via adsorption at the metal/solution interface. The mechanism by which an inhibitor decreases the corrosion current is achieved by interfering with some of the steps for the electrochemical process. The corrosion inhibition of mild steel in aggressive acidic solutions has been widely investigated [4-27]. In industries, hydrochloric acid solutions are often used in order to remove scale and salts from steel surfaces, and cleaning tanks and pipelines. This treatment may be prerequisite for coating by electroplating, galvanizing, or painting techniques. The acid must be treated

to prevent an extensive dissolution of the underlying metal. This treatment involves the addition of some organic inhibitors to the acid solution that adsorb at the metal/solution interface by displacing water molecules on the surface and forming a compact barrier film. Nitrogen-containing compounds as metal corrosion inhibitors have been studied thoroughly by many authors. These compounds include benzimidazole and imidazole derivatives [7, 8], bipyrazole [26], stilbazole [27], quaternary ammonium salts [28-35], polyaminobenzoquinone polymers [36], substituted aniline-N-salicylidenes [37], amides [38], heterocyclic compounds [39, 40], and cationic surfactants [41, 42]. Other authors worked on phosphorous containing and sulphur-containing inhibitors [43-45]. Other studies involved the effect of addition of some ions on the inhibition efficiency of some organic compounds [46, 47]. In all these studies, the nitrogen, phosphorous, and sulphur atom(s) in the compounds as well as the unsaturated double bonds like phenyl rings showed to be able to adsorb very well on the metal surface and form protective layer, which in turn increased the corrosion inhibition with the increase in the concentration of the inhibitor, in some cases reaching 99% inhibition.

This work is aimed at determining the inhibition effect of *Aspilia africana* on mild steel corrosion in 1.0 M HCl solution. Mild steel was chosen in our studies since high temperature aggressive acids are widely used in industries in connection to mild and low alloy steels. This work includes using gravimetric measurements and the effect of temperature on the corrosion inhibition of mild steel in 1.0 M HCl solution to calculate the thermodynamic parameters.

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2. Experimental

2.1. Mild Steel Preparation

Mild steel specimens (C = 0.08 wt.%, Si = 0.05 wt.%, P = 1.00 wt.%, Cu = 0.02 wt.%, Pb = 0.02 wt.% and Fe = 98.83 wt.%) of dimensions of 20 x 20 x 1 mm were used for the gravimetric study. The surface preparation of mechanically polished specimens was carried out using different grades of emery paper and then degreased with acetone and air-dried.

2.2. Preparation of *A. africana* Extract

A. africana leaves were collected around Umuahia, Nigeria. The leaves were cleaned, dried at room temperature, ground, and 4 grams of the ground leaves were soaked in distilled water. The crude extracts were boiled at 75°C in reflux apparatus for 3 hours, allowed to cool, and filtered. After filtration, a 198ml stock (extract) solution containing 1.09g of the leaves was obtained. The amount of ground leaves material extracted into solution was quantified by comparing the weight of dried residue with initial weight of the dried leaf material before extraction. From the respective stock solutions, inhibitor test solution was prepared in 5 concentrations ranged from 0.1 to 0.5 g/L.

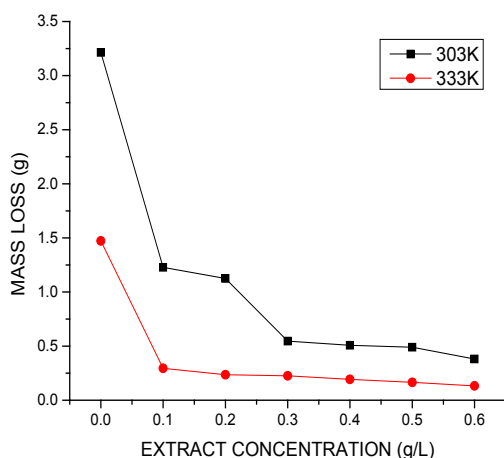


Figure 1. Mass Loss Values of Mild Steel corrosion in 1 M HCl in the Absence and Presence of *Aspilia africana* Leaves Extract after 72 hours Exposure Time at 303K and 333K

2.3. Gravimetric Technique

The polished and pre-weighed mild steel specimens of uniform size were suspended in 300 ml test solutions with and without the inhibitor at different concentrations for a period of 40 h. The experiment was repeated in a water bath set at 60°C (333K). Then, the specimens were washed, dried and weighed. The mass-loss was calculated. From these data, inhibition efficiency (I%) was calculated using the following equation:

$$I\% = \left(1 - \frac{\rho_1}{\rho_0}\right) \times 100 \quad (1)$$

Where ρ_0 is the corrosion rate of mild steel without

inhibitor and ρ_1 is the corrosion rate of mild steel with inhibitor. Corrosion rate (ρ) is calculated from the equation:

$$\rho = \frac{K\Delta W}{ADt} \quad (2)$$

where, K is the year constant, ΔW is the mass-loss (in g), A is the surface area of the specimen (in cm²), D is the density (in g/cm³), t is the period of exposure (in hours).

3. Results and Discussion

3.1. Gravimetric Studies

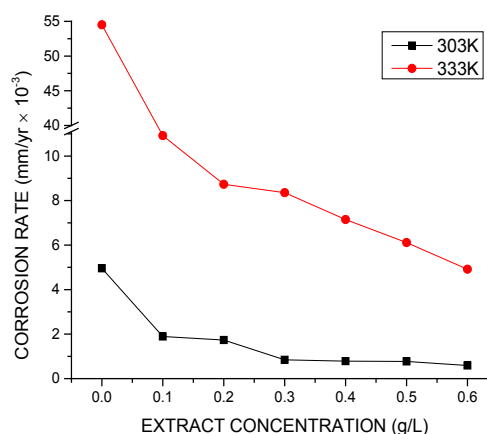


Figure 2. Corrosion Rate Values of Mild Steel corrosion in 1 M HCl in the Absence and Presence of *Aspilia africana* Leaves Extract after 72 hours Exposure Time at 303K and 333K

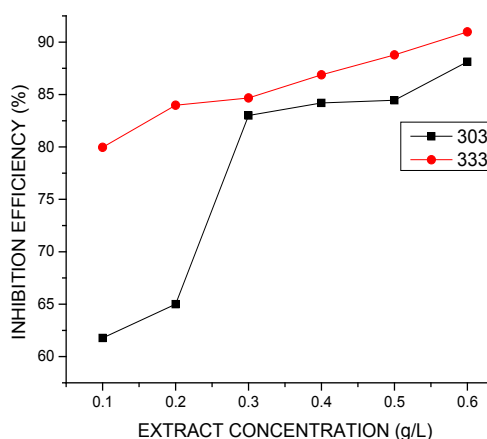


Figure 3. Inhibition Efficiency of *Aspilia africana* Leaves Extract on Mild Steel corrosion in 1 M HCl after 72 hour Exposure Time at 303K and 333K

The results of the gravimetric studies are shown in figures 1-3 below. Figure 1 shows the mass loss plots of the corrosion of mild steel in the absence and presence of *Aspilia africana* leaves extract which depict that, at both temperatures, the mass of the metal was reduced with introduction of the extract and the inhibition improved with increase in inhibitor concentration. Figure 2 shows the

corrosion rate values calculated from the values of the uninhibited and inhibited corrosion of the mild steel specimens, and Figure 3 shows the inhibition efficiency values of the different concentrations of *Aspilia africana* leaves extract on mild steel corrosion. The results depict optimum inhibition efficiency of *Aspilia africana* leaves extract of about 88.1% at 303K and 91% at 333K. At both temperature conditions, the corrosion rate (Figure 2) was rapidly reduced with introduction of the inhibitor which became less rapid with increase in concentration of inhibitor except for the third concentration which was slightly rapid for the 303K temperature and less rapid in the 333K temperature. It was observed that the rate of deterioration of mild steel reduced significantly at increased temperature in the presence of the inhibitor. The inhibitive effect of the inhibitor is attributed to the phytochemical constituent of the plant leaves especially the sulphur, nitrogen and oxygen heteroatoms. It can be suggested that the inhibitor molecules present in the leaves extract block the surface of mild steel via adsorption mechanism. The higher but slow rate of corrosion with increase in temperature can be attributed the chemical adsorption of the metal which takes more time than in physisorption.

3.2. Adsorption Mechanism

Langmuir isotherm was used to deduce whether there is formation of layer of insoluble complex of the metal on the surface which acts as a barrier between the metal surface and the corrosive medium - usually termed physisorption, while Temkin isotherm was used to determine if the extract adsorption on the metal surface is via chemisorption which involves displacement of water molecules from the metal surface and the sharing of electrons between oxygen atom and iron. Langmuir plot describes the relationship between the surface coverage and inhibition concentration of a material as;

$$\frac{C}{\theta} = \frac{1}{K} + C \quad (3)$$

Temkin isotherm plot is expressed as;

$$\theta = \left(\frac{1}{f}\right) \ln(KC) \quad (4)$$

where C is the concentration of inhibitor, θ is the surface coverage, f determines the adsorbent-adsorbates interaction and K is the equilibrium constant employed in calculating the Gibb's free energy (Equation 5) [48-51]. However, Temkin adsorption isotherm assumes a uniform distribution of adsorption energy, which decreases with the increase of the value of surface coverage, θ . It focuses on the chemisorptions aspect of corrosion inhibition of organic plants on metals. Figure 4 shows Langmuir adsorption isotherm plot with a straight line curve with the slopes (1.0175 and 1.0724) and correlation coefficients (0.989 and 0.999) being about unity. Figure 5 also shows a straight line curve for the Temkin adsorption isotherm plot with the slopes vanishing (0.1603 and 0.0573) and correlation coefficients (between 0.856 and 0.942) being about unity.

$$\Delta G_{ads}^{\circ} = -RT \ln 55.5K \quad (5)$$

where R is the gas constant (8.314kJ/mol) and T is the temperature in Kelvin. 55.5 is the concentration of water in solution in mol/L. The negative value of ΔG_{ads}° indicates spontaneity in the inhibitor adsorption on mild steel surface.

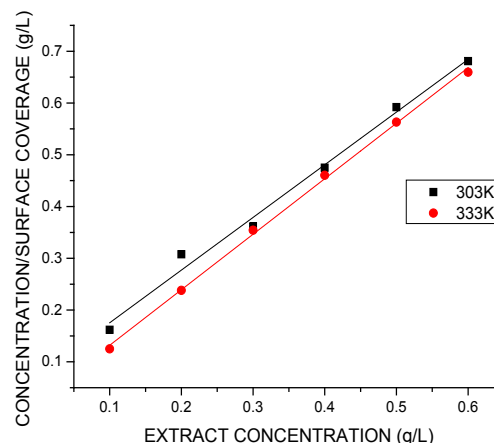


Figure 4. Langmuir Isotherm Plots of the Corrosion of Mild Steel corrosion in 1 M HCl in the Presence of *Aspilia africana* at 303K and 333K

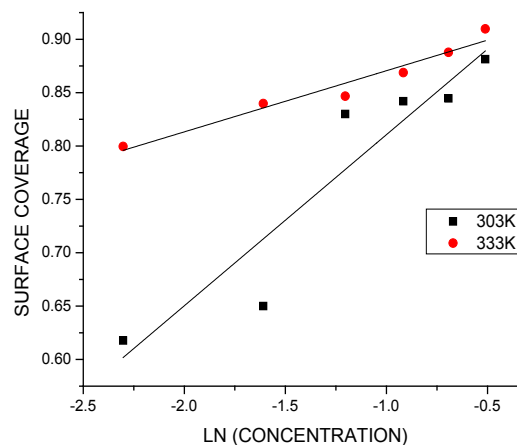


Figure 5. Temkin Isotherm Plots of the Corrosion of Mild Steel corrosion in 1 M HCl in the Presence of *Aspilia africana* at 303K and 333K

3.3. Effect of Temperature

Analysis of the temperature dependence of inhibition efficiency as well as comparison of corrosion apparent activation energies in absence and presence of inhibitor gives some insight into the possible mechanism of inhibitor adsorption. A decrease in inhibition efficiency with the rise in temperature with analogous increase in corrosion activation energy in the presence of inhibitor compared to its absence is frequently interpreted as being suggestive of formation of an adsorption film of physical (electrostatic) nature. The reverse effect, corresponding to an increase in inhibition efficiency with rise in temperature and lower activation energy in the presence of inhibitor, suggests a chemisorptions mechanism [47, 52, 53].

Table 1. Adsorption parameters obtained from temperature and isotherm plots of the corrosion rate values of mild steel in *A. africana*

ISOTHERM		SLOPE	INTERCEPT	K	R ²	ΔG [°] _{ads} (kJ/mol)
LANGMUIR	303K	1.0175	0.07369	13.57	0.989	-16.69
	333K	1.0724	0.02477	40.37	0.999	-21.36
TEMKIN	303K	0.1603	0.97094	427.10	0.856	-25.38
	333K	0.0573	0.92785	1.08 × 10 ⁷	0.942	-55.96

In examining the effect of temperature in the corrosion process in the presence of the extracts, the Arrhenius equation is used [54]:

$$\log \frac{\rho_2}{\rho_1} = \frac{E_a}{2.303R} \left(\frac{1}{T_1} - \frac{1}{T_2} \right) \quad (6)$$

where E_a is the apparent activation energy, R is the molar gas constant, T is the absolute temperature, A is the Arrhenius constant, ρ_1 and ρ_2 are the corrosion rates at temperatures T_1 and T_2 respectively. θ_1 and θ_2 are the degrees of surface coverage at temperatures T_1 and T_2 . Decreased values of E_a from the non-inhibited solution indicate that the inhibition efficiency of the extract is greater at increased temperature, while increased values of E_a indicate that the inhibition efficiency of the extract is reduces at increased temperature.

Table 2. Calculated values of apparent activation energy (E_a) and heat of adsorption (Q_{ads}) of *Aspilia africana* leaves extract at 303K and 333K temperatures

EXTRACT CONCENTRATION	E_a (kJmol ⁻¹)	Q_{ads} (kJmol ⁻¹)
0	67.05	--
0.10	49.00	25.27
0.20	45.20	29.02
0.30	64.16	3.45
0.40	61.86	6.08
0.50	57.94	10.50
0.60	59.37	8.57

Estimates of the heats of adsorption (Q_{ads}) were obtained from the trend of surface coverage with temperature as follows [55]:

$$Q_{ads} = 2.303R \left[\log \left(\frac{\theta_2}{1-\theta_2} \right) - \log \left(\frac{\theta_1}{1-\theta_1} \right) \right] \times \frac{T_1 T_2}{T_2 - T_1} \quad (7)$$

A positive Q_{ads} value signifies that the degree of surface coverage increased with rise in temperature. Such behaviour is often interpreted as being indicative of chemical adsorption of organic matter on the steel surface [56], but the tendency to negativity signifies physical adsorption mechanism.

4. Conclusions

This study shows that *Aspilia africana* leaves extract is a good inhibitor of mild steel corrosion in 1.0M HCl at room temperature and at 60°C. The values the Gibb's free energy

of adsorption indicates that *Aspilia africana* leaves extract were chemically and physically adsorbed on the surface of the metal. The values of the slopes being about unity for the Langmuir plots and also R^2 value is indicative of Physical adsorption, while the vanishing slope of the Temkin plots and R^2 value about unity signifies chemical adsorption. The deductions favour mixed inhibition. This is further confirmed from the fluctuating values obtained from the calculations of the apparent activation energy and heat of adsorption which also depict a mixed. Thus *Aspilia africana* leaves extracts can be used in the acid cleaning of mild steel to reduce the rate of corrosion of the metal.

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