Corrosion Inhibitive Properties and Adsorption Behaviour of Acetone Extract of *Anacardium occidentale* on Mild Steel and Aluminium in HCl

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The corrosion inhibition of aluminium and mild steel in hydrochloric acid by acetone extract of *Anarcardium occidentale* was studied using the weight loss technique at 28 ºC. The result of the study reveals that the different concentrations of the extract of *Anarcardium occidentale* inhibit aluminium and mild steel corrosion. The inhibition efficiency and surface coverage increases with an increase in the concentration of the extract. Thermodynamic consideration reveals that the adsorption on mild steel and aluminium surface is spontaneous and occurs according to Temkin and Freundlich adsorption isotherms. The phenomenon of physical adsorption is proposed from the free energies ΔG°_{ads} *i.e.* the ΔG°_{ads} is > 40 KJ/mol and negative; suggestive of adsorption on the metal. The ΔG° for mild steel is -7.695 KJ/mol and -9.23 KJ/mol for aluminium. A first order type of mechanism has been deduced from the kinetic treatment of the results and values of some thermodynamic parameters were evaluated and compared with reference to mild steel and aluminium.

Key Words: Adsorption, Corrosion, Inhibitors, *Anacardium occidentale*, Mild steel, Aluminium.

INTRODUCTION

In various manufacturing industries and firms, the need to use constructional materials safely, but cost effectively is a primary focus. Metals like aluminium, iron and their alloys are of utmost importance in constructional and metallurgical work.

Aluminium and its alloys have a remarkable economic and attractive material for engineering applications owing to its low cost, light weight, high thermal and electrical conductivity[1]. It is widely used as a corrosion resistant metal but in the presence of aggressive ions such as chloride ion, the protective layer can be locally destroyed and corrosion attack takes place[2].

On the other hand, mild steel is one of the most important widely used engineering materials particularly for the structural, functional and metallurgical applications. Unlike aluminium, mild steel undergoes rusting easily in the humid atmosphere (presence of oxygen and water). Hence, the corrosion of aluminium and its alloy, mild steel and other metals in various environment (neutral, acidic, alkaline) have been a continuous research.

In an attempt to checkmate corrosion, a variety of inhibitors have been employed which are either organic or inorganic in nature. Most of the well known acidic inhibitors are organic compounds containing electron donor atoms particularly nitrogen, sulfur, oxygen in their functional group with aromatic and heterocyclic rings[3]. Nitrogen and oxygen are more effective corrosion inhibitors in acidic media. Inorganic substances such as phosphates, chromates (hexavalent chromium) and arsenates have been found as effective metal corrosion inhibitor, but a major setback is their toxicity and as such, their use has come under severe scrutiny and criticisms.

Currently, researches are geared towards finding suitable alternative corrosion inhibitors to replace the inorganic and organic compounds. Naturally occurring substances like plant exudates have been found to readily satisfy this need. Apart from being readily available, cheap and a renewable source of material, they are eco-friendly and ecologically acceptable[4].

In the quest to find an alternative and more importantly an effective, less toxic, less expensive, ecologically friendly corrosion inhibitor to organic and inorganic inhibitors, tannin extract from the testa of *Anacardium occidentale* is being proposed. Hence, this work studies the use of gravimetric weight loss method in investigating the corrosion inhibition of mild steel and aluminium using tannin from *Anacardium occidentale* (cashewnut testa) in HCl at 28 ºC.

EXPERIMENTAL

Preparation of the cashewnut testa: The cashew nut testa(s) was collected from cashew nut sellers at Ogige market, Nsukka. Impurities like cashew seeds, gravel and sand were

removed from the testa by winnowing and sieving. Then the cashew testa were subjected to a size reduction mechanisms into a partially ground particles. Ground cashew nut testa (200 g) were soaked with 70 % acetone in a 5 L plastic bowl and allowed to stand for 2 days with intermittent stirring. The filtrate containing acetone was distilled over at 50 ºC to recover the acetone. The resultant tannin extract was then quantified spectrophotometrically using the Folin-Ciocalteau method.

Preparation of the tannic acid standard/calibration curve: Folin-Ciocalteau reagent (0.2 mL) was added to tannic acid (5 mL); a bluish black colouration forms which was allowed to stand for 5 min. 25 % Na_2CO_3 (4 mL) solution was added to the solution. After 20 min, the absorbances were read at 700 nm against a reagent blank using a B. Bran 700-2000 spectronic 20D spectrophotometer. The absorbance values were plotted against concentration to obtain the standard curve (a Beer-Lambert plot).

Tannin extract: Folin-Ciocalteau reagent (0.2 mL) was added to the (10-200 ppm) tannin extract (5 mL) with a dilution factor of (1:1000). The bluish black solution was allowed to stand for 5 min before adding 25 % Na_2CO_3 solution (4 mL) to the solution. After 20 min the absorbance values were read at 700 nm against a reagent blank. The absorbance values of the tannin extract were compared with the calibration curve. Extrapolation of the absorbance values from the tannic acid standard curve (Fig. 1), gave the concentration of the tannin extract.
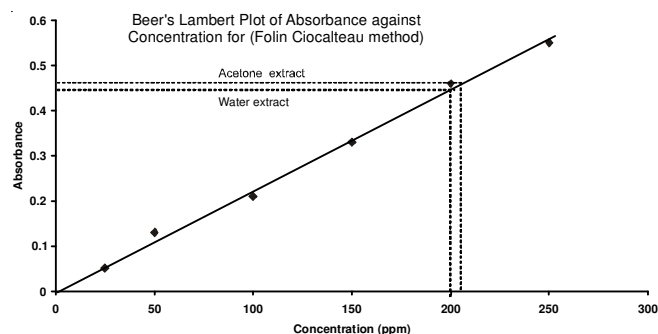

Fig. 1: Tannic acid standard curve (Folin-Ciocalteau method)

Preparation of the metal coupons: The mild steel and aluminium with thickness of 0.2 cm and 0.1 cm, respectively were obtained from a mechanical workshop along Enugu Road, Nsukka and mechanically pressed-cut approximately into 3.2 cm × 2.5 cm (mild steel) and 3.5 cm × 2.5 cm (aluminium) coupons. These coupons were used without further polishing. However, for surface treatment, they were degreased in absolute ethanol, washed in distilled water, dried in acetone and stored in a moisture free desicator before use in the corrosion studies.

Preparation of the corrosion media

Three different media were prepared: (i) One containing the tannin extracts and the corrodent HCl made up to 50 cm³. The tannin is varied from (1.0-1.5 g) while the concentration of HCl is maintained at (1 M). (ii) Another containing both tannin extract and HCl where the tannin is kept constant (1 g) while the concentration of HCl is varied from (0.01-1.50 M) and made up to 50 cm³. (iii) A control experiment containing

only HCl (0.01-1.50 M) alone with no acetone extract of *Anarcardium occidentale* in it.

Procedure: The pre-cleaned weighed aluminium and mild steel coupons were immersed completely in a 250 mL plastic bowl(s) containing HCl and acetone extract of *Anarcardium occidentale*. The experiment was at a constant temperature of 28 ºC. The coupons were retrieved at 24 h interval progressively for 168 h (7 days), washed in distilled water and dried in acetone and weighed. The differences in weight of the coupons were calculated using the formula:

$$W = W_o - W_t \qquad (1)$$

where W_o, is the initial weight of the coupons before immersion in the corrosion media (g), W_t, is the final weight of the coupons after immersion in the corrosion media (g) and W is the total weight loss (g). Each reading reported is an average of three experimental reading recorded to the nearest 0.001 g.

The inhibition efficiency of acetone extract of *Anarcardium occidentale* acting as inhibitor was calculated using the formula:

$$IE\ (\%) = \left[\frac{W_2 - W_1}{W_2} \right] \times \frac{100}{1} \qquad (2)$$

where W_1 and W_2 are the weight losses for mild steel and aluminium coupons in the absence and presence of the inhibitor respectively in HCl solution at 28 ºC.

The degree of surface coverage, θ is given by the equation:

$$\theta = 1 - \frac{W_1}{W_2} = \frac{W_2 - W_2}{W_2} \qquad (3)$$

The corrosion rate (CR) of mild steel and aluminium in the different corrosion medium was determined for a 168 h immersion period from weigh loss using the formula:

$$\Delta W/AT \qquad (4)$$

where, $\Delta W = (W_1 - W_2)$ (g), A is the area of the metal coupons (cm³) and T is the exposure time (h).

RESULTS AND DISCUSSION

Effect of corrodent concentration on mild steel and aluminium corrosion: The corrosion of aluminium and mild steel in different concentration of HCl (0.01-1.50 M HCl) as corrodent at 28 ºC without inhibitor is shown in Fig. 2. This shows a weight loss concentration curve of mild steel and aluminium dissolution in HCl in the absence of acetone extract of *Anarcardium occidentale*. The result obtained indicates that weight loss of aluminium and mild steel increased with increasing HCl concentration.

Furthermore, the corrosion of the metal coupons increases linearly with increasing time as shown in Fig. 3 (weight loss time curve). It was observed that the weight loss of mild steel was greater than that of aluminium at lower concentration of HCl (from 0.01-0.50 M). But at higher concentration of HCl (1.0-1.5 M), the weight loss of aluminium was higher than that of mild steel.

The corrosion can be attributed to the fact that the rate of chemical reaction increases with increasing H^+ concentration or probably due to increase in the rate of diffusion and ionization
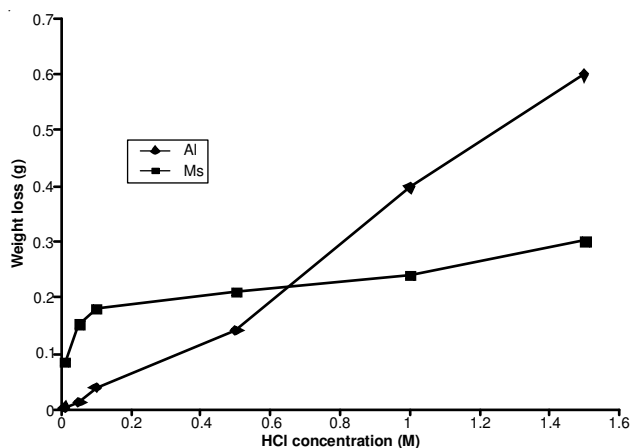
Fig. 2. Plot of weight loss for mild steel and aluminium against concentration of HCl alone at 28 ºC for t = 7 days
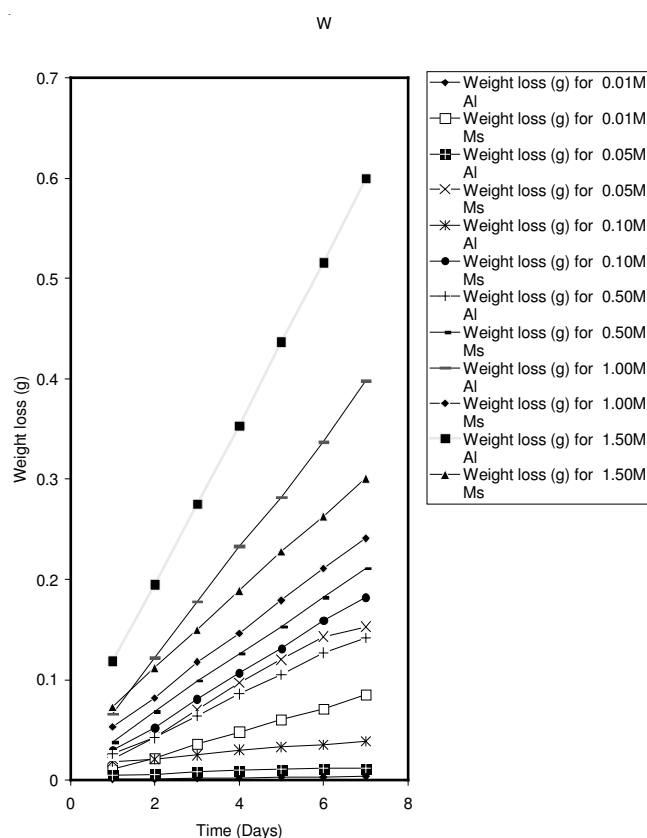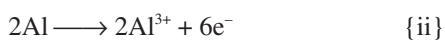

Fig. 3. Plot of weight loss against time for aluminium and mild steel in varied HCl concentration alone at 28 ºC
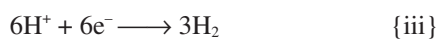
of active species in the corrosion reaction and/or probably due to passivity as in the case of aluminium[5]. Aluminium is rapidly corroded by HCl according to the equation below:

$$2Al + 6HCl \longrightarrow 2AlCl_3 + 3H_2 \qquad \{i\}$$

The oxidation reaction is:

$$2Al \longrightarrow 2Al^{3+} + 6e^- \qquad \{ii\}$$

The reduction reaction is:

$$6H^+ + 6e^- \longrightarrow 3H_2 \qquad \{iii\}$$

The general ionic equation is:

$$2Al + 6H^+ + 6e^- \longrightarrow 2Al^{3+} + 3H_2 + 6e^- \qquad \{iv\}$$
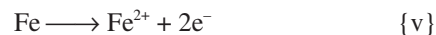
As a result of these reactions including the high solubility of the corrosion products, the aluminium metal losses weight in HCl solution[2].

On the other, the dissolution of mild steel in HCl is as follows:

$$Fe \longrightarrow Fe^{2+} + 2e^- \qquad \{v\}$$

The dissolution is initiated at the surface of the metal, which is the reaction site.

Effects of the inhibitor concentration on the corrosion of mild steel and aluminium metal coupons: The corrosion of aluminum and mild steel in different concentration of HCl as corrodent and tannin of *Anacardian occidentale* as inhibitor is shown in Figs. 4 and 5.
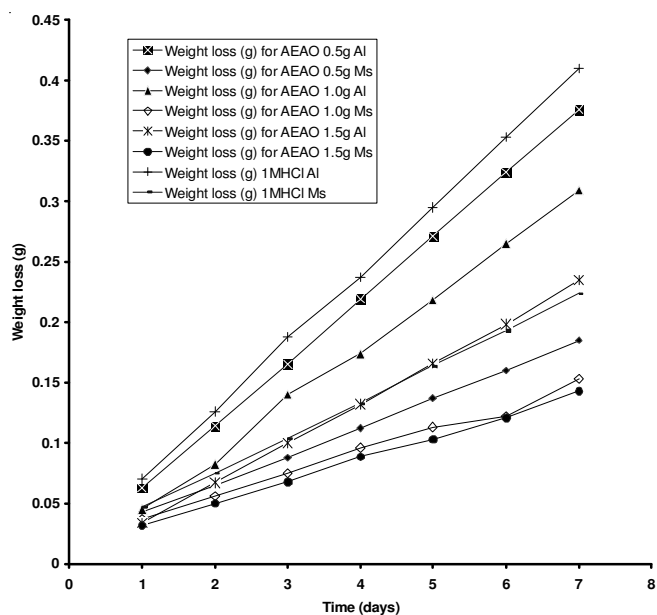

Fig. 4. Plot of weight loss for mild steel and aluminium against time for varied HCl concentration in 1 g acetone extract of *Anarcardium occidentale* at 28 ºC
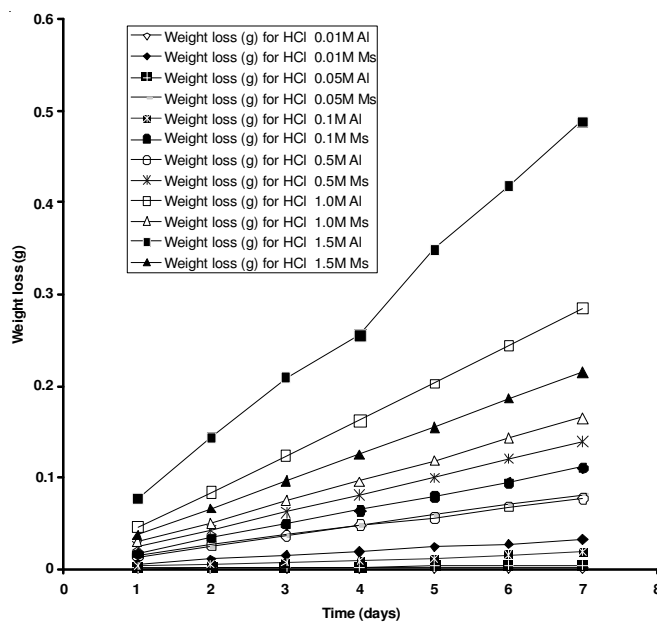

Fig. 5. Plot of weight loss for aluminium and mild steel against time for varied HCl concentration in 1 g acetone extract of *Anarcardium occidentale* at 28 ºC

The weight loss concentration and time curve respectively reveal that acetone extract of *Anarcardium occidentale* inhibit the corrosion of aluminium and mild steel in different HCl concentration since there was a general decrease in weight loss. This decrease is lower than the corresponding weight loss of the metal coupons in HCl solution alone. However, corrosion was higher in aluminium than mild steel in the presence of different concentration of acetone extract of *Anarcardium occidentale* in 1 M HCl.

It is of general view that the inhibition of metals in acidic medium results from the adsorption of molecules or ions of the inhibitor on the metal surface. The action of organic inhibitors depends on the type of interaction between the substance and the metallic surface.

The behaviour of the inhibitor extracts also indicates that the adsorbed molecules of the inhibitor actually form a hydrophobic film which screened the metal surfaces and therefore protects the surfaces from the action of the corrosive medium[2].

The inhibition efficiency values were calculated using eqn. 2. From Table-1, it is clearly seen that the inhibition efficiency increases in the presence of the acetone extract of *Anarcardium occidentale*.

TABLE-1
VARIATION OF AEAO CONCENTRATION AND INHIBITION EFFICIENCY FOR MILD STEEL AND ALUMINIUM IN 1 M HCl AT 28 ℃

Conc. of AEAO (g)	IE (Mild steel) (%)	IE (Aluminium) (%)
0.5	8.30	17.40
1.0	24.60	31.7
1.5	42.70	36.2

AEAO = Acetone extract of *Anarcardium occidentale*

Furthermore, the inhibition efficiency was observed from the plot in Fig. 6, to increase with increased acetone extract of *Anarcardium occidentale* concentration.
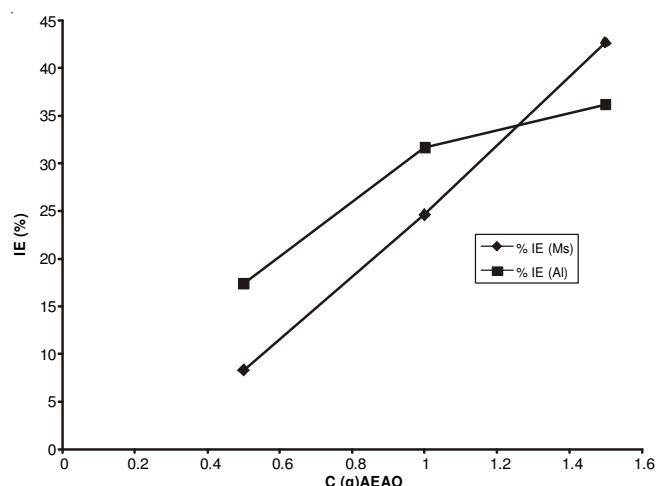

Fig. 6. Plot Inhibition Efficiency of mild steel and aluminium at constant 1 M HCl, against acetone extract of *Anarcardium occidentale* concentration at 28 ℃

For 1 M HCl and varied acetone extract of *Anarcardium occidentale* mixture, the maximum and minimum inhibition efficiency was 42.7 and 8.3 % for the aluminium coupons. At 0.5 g and 1.0 g acetone extract of *Anarcardium occidentale*

concentration, the inhibition efficiency of mild steel were quite higher (17.4 and 31.7 %, respectively) than that of the aluminium (8.3 and 24.6 %) (Table-1). However aluminium has the highest inhibition efficiency (42.7 %).

For the 1 g acetone extract of *Anarcardium occidentale* (constant) and varied concentration of HCl, the maximum and minimum inhibition efficiency were 75 and 18.5 % for aluminium at 0.01 M and 1.5 M HCl and 62.4 and 28.6 % for mild steel at 0.01 M and 1.5 M HCl, respectively (Table-2). This further shows that the IE % of aluminium is higher than that of mild steel. This is also true for the surface coverage θ (the calculated values of θ are given in Tables 3 and 4).

TABLE-2
INHIBITION EFFICIENCY OF MILD STEEL AND ALUMINIUM WITH VARIED HCl CONCENTRATION IN 1 g ACETONE EXTRACT OF *Anarcardium occidentale* AT 28 ℃

S/N	Conc. of HCl (M)	IE (Mild steel) (%)	IE (Aluminium) (%)
1	0.01	62.40	75.00
2	0.05	46.10	66.70
3	0.10	39.00	51.30
4	0.50	34.10	45.80
5	1.00	31.50	26.70
6	1.50	28.60	18.50

TABLE-3
SURFACE COVERAGE θ AND CORROSION RATE (CR) IN gh^{-1} cm^{-3} VARIATION FOR MILD STEEL AND ALUMINIUM WITH VARIED AEAO CONCENTRATION IN 1 M HCl AT 28 ℃

C (g)	θ Al	θ Ms	CR (Al) gh^{-1} cm^{-3}	CR (MS) gh^{-1} cm^{-3}
0.50	0.083	0.174	2.31×10^{-4}	1.45×10^{-4}
1.00	0.246	0.317	6.87×10^{-4}	2.60×10^{-4}
1.50	0.427	0.362	1.19×10^{-3}	3.01×10^{-4}

C = concentration of acetone extract of *Anarcardium occidentale* (g), MS = Mild steel, Al = Aluminium

TABLE-4
SURFACE COVERAGE θ AND CORROSION RA1E (CR) IN gh^{-1}cm^{-3} VARIATION OF MILD STEEL AND ALUMINIUM WITH VARIED HCl CONCENTRATION AND 1 g ACETONE EXTRACT OF *Anarcardium occidentale* CONCENTRATION AT 28 ℃

C (M)	θ Al	θ Ms	CR (Al) gh^{-1} cm^{-3}	CR (MS) gh^{-1} cm^{-3}
0.01	0.750	0.624	2.04×10^{-5}	1.97×10^{-4}
0.05	0.667	0.461	5.44×10^{-5}	2.60×10^{-4}
0.10	0.513	0.390	1.36×10^{-4}	2.64×10^{-4}
0.50	0.458	0.341	4.42×10^{-4}	2.68×10^{-4}
1.00	0.267	0.315	7.07×10^{-4}	2.83×10^{-4}
1.50	0.185	0.286	7.55×10^{-4}	3.20×10^{-4}

C = Concentration of HCl in molarity (M), MS = Mild steel, Al = Aluminium

The corrosion rate calculations from eqn. 4 indicate that the metal coupons weight loss decreases with increase in the concentration of the inhibitor (acetone extract of *Anarcardium occidentale*) and increases with decreases in the concentration of the inhibitor (acetone extract of *Anarcardium occidentale*). This is graphically shown in Figs. 8 and 9. The calculated values of the corrosion rate of mild steel and aluminium are also shown in Tables 3 and 4.
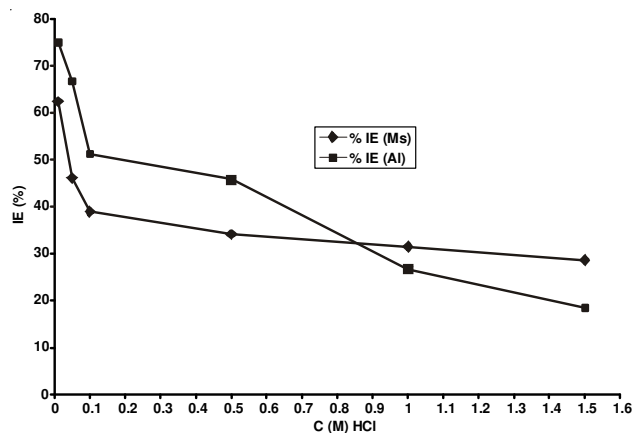
Fig. 7.   Plot of Inhibition efficiency of Mild steel and aluminium with varied HCl concentration in 1 g acetone extract of *Anarcardium occidentale* at 28 ºC
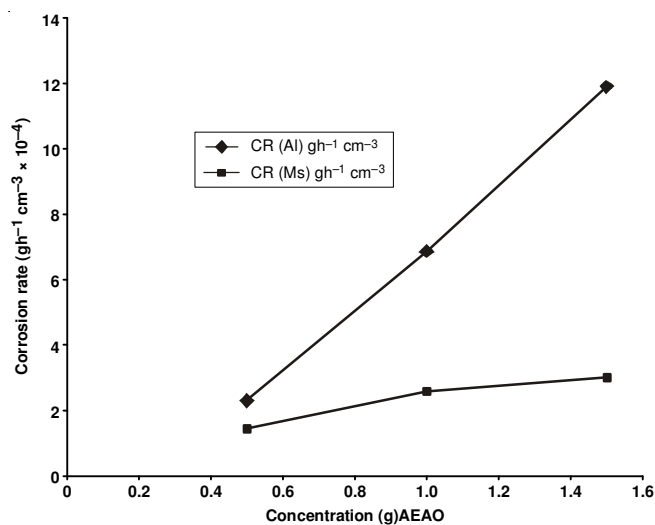

Fig. 8.   Plot of corrosion rate for mild steel and aluminium with varied concentration of acetone extract of *Anarcardium occidentale* in 1 M HCl at 28 ºC
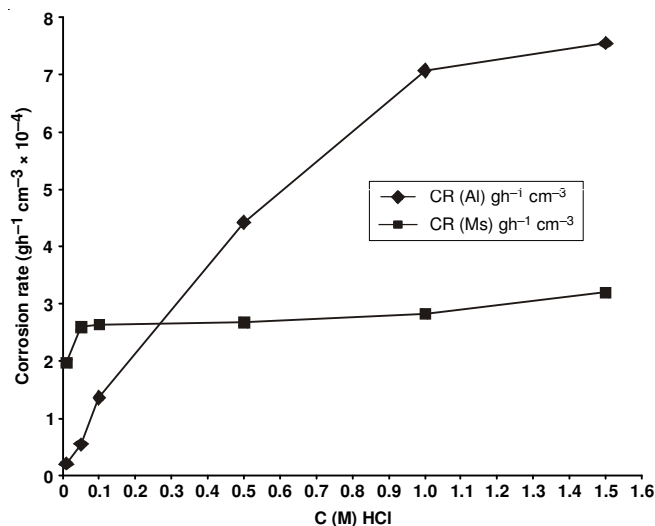

Fig. 9.   Plot of corrosion rate for mild steel and aluminium with varied concentration of HCl in 1 g acetone extract of *Anarcardium occidentale* at 28 ºC

Kinetic analysis of weight loss results: Corrosion reaction is an overall reaction in which both solid and liquid phases are consumed. It is therefore difficult to apply most of the principles of chemical kinetics to corrosion reactions.

Plots of log W_f against time, t in days at 28 ºC in HCl for different systems in the presence and absence of acetone extract of *Anarcardium occidentale* studies reveals linear plots signifying first-order kinetics as depicted graphically in Figs. 10-13. This is in agreement with works done by other authors[6-8].

The linear relation is given as:

$$\log W_f = \log W_o - K_t \qquad (5)$$

where W_f and W_o are the weight of coupons after (final weight loss) and before (initial weight loss) immersion respectively, k is the rate constant and t is time.

The values of the rate constants were obtained from the slope in Figs. 9-12.

From the value of the rate constant, half life, $t_{1/2}$ of corrosion reaction involving aluminium and mild steel metal coupons in the presence and absence of different concentration of acetone extract of *Anarcardium occidentale* were calculated.
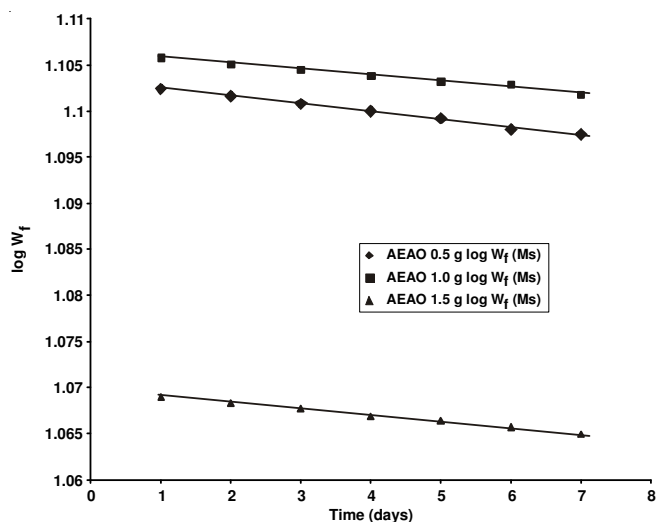

Fig. 10. Plot of log W_f for mild steel with varied acetone extract of *Anarcardium occidentale* concentration against time in 1 M HCl at 28 ºC
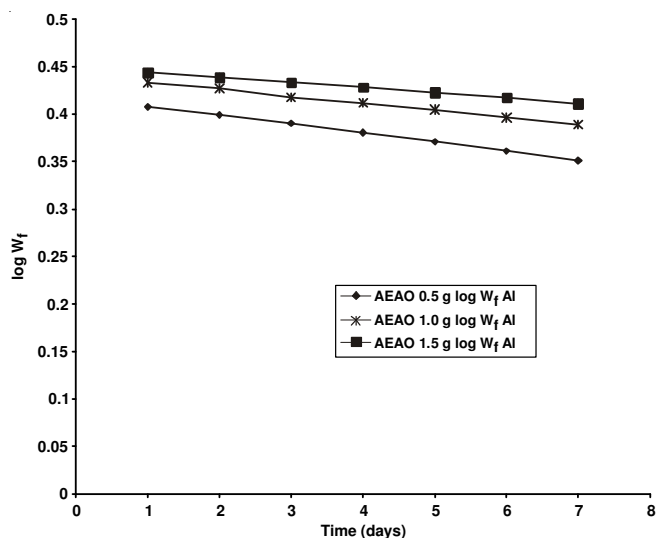

Fig. 11. Plot of log W_f for aluminium with varied acetone extract of *Anarcardium occidentale* concentration against time in 1 M HCl at 28 ºC
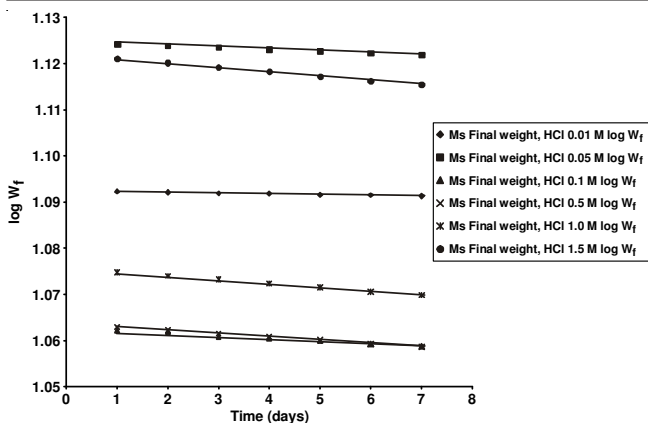
Fig. 12. Plot of log W_f for mild steel with varied HCl concentration in 1 g acetone extract of *Anarcardium occidentale* concentration against time at 28 ℃
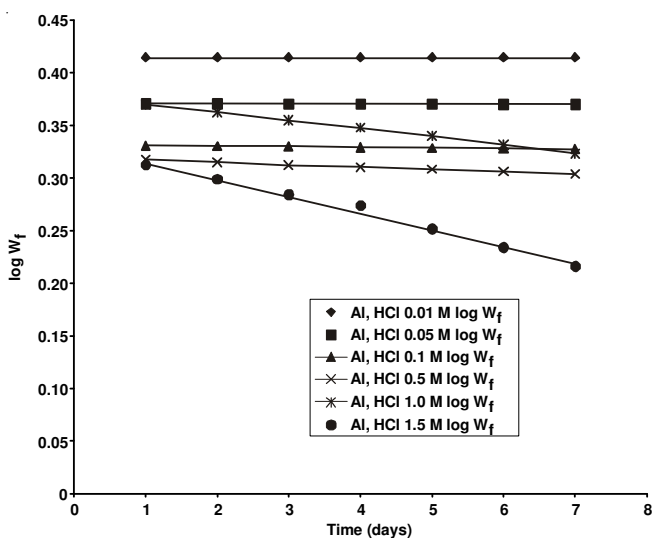

Fig. 13. Plot of log W_f for aluminium with varied HCl concentration in 1 g acetone extract of *Anarcardium occidentale* concentration against time at 28 ℃

The $t_{1/2}$ for first order is given as:

$$0.693/k \qquad (6)$$

where k is the rate constant.

The values of k and $t_{1/2}$ are presented in Tables 5 and 6.

At constant 1M HCl concentration and varied acetone extract of *Anarcardium occidentale* concentration, the $t_{1/2}$ was increasing as the concentration of acetone extract of *Anarcardium occidentale* increases, with mild steel having a higher $t_{1/2}$ value. This is depicted in Fig. 14.

On the other hand, when the concentration of HCl was varied and acetone extract of *Anarcardium occidentale* maintained at 1 g, the $t_{1/2}$ was found to decrease with increasing concentration

TABLE-5
RATE CONSTANT AND HALF-LIFE VARIATION FOR MILD STEEL AND ALUMINIUM WITH VARIED ACETONE EXTRACT OF *Anarcardium occidentale* CONCENTRATION IN 1 M HCl AT 28 ℃

C (g)	k (Al) days⁻¹	k (Ms) days⁻¹	$t_{1/2}$ (Al) days	$t_{1/2}$ (Ms) days
0.5	9.450×10^{-3}	8.165×10^{-4}	73.3	848.7
1.0	7.357×10^{-3}	6.611×10^{-4}	94.2	1048.3
1.5	5.434×10^{-3}	6.887×10^{-4}	127.5	1006.2

TABLE-6
RATE CONSTANT AND HALF-LIFE VARIATION FOR MILD STEEL AND ALUMINIUN WITH VARIED HCl CONCENTRATION IN 1 g ACETONE EXTRACT OF *Anarcardium occidentale* AT 28 ℃

C (M)	k (Al) days⁻¹	k (Ms) days⁻¹	$t_{1/2}$ (Al) days	$t_{1/2}$ (Ms) days
0.01	2.78×10^{-5}	1.52×10^{-4}	24892.2	4559.2
0.05	9.25×10^{-5}	3.76×10^{-4}	7491.9	1843.1
0.10	5.43×10^{-4}	5.92×10^{-4}	1276.2	1170.6
0.50	2.20×10^{-3}	7.17×10^{-4}	315.0	966.0
1.00	7.78×10^{-3}	8.27×10^{-4}	89.0	837.6
1.50	1.60×10^{-2}	9.54×10^{-4}	43.3	726.4

of HCl (Fig. 15). The rate constant increases as the concentration of HCl increases (Fig. 16).

Adsorption consideration: The mechanism of the interaction between inhibitor and the metal surface can be explained using adsorption isotherms. All these isotherms are of the general form:

$$f(\theta, x) \exp(-2a\theta) = KC \qquad (7)$$
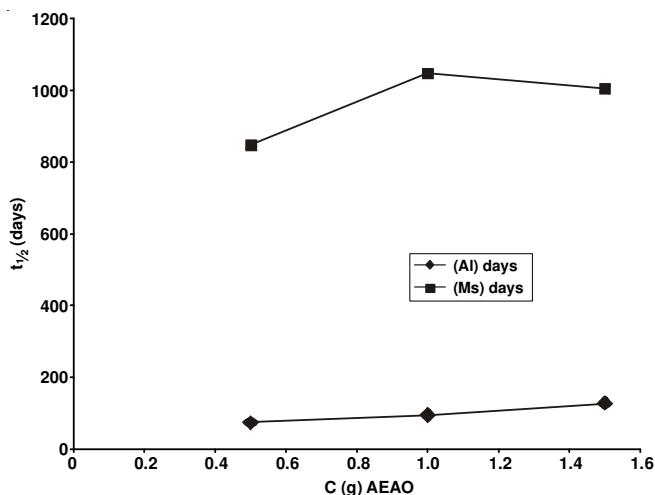

Fig. 14. Plot of half life for mild steel and aluminium against varied acetone extract of *Anarcardium occidentale* concentration in 1 M HCl at 28 ℃
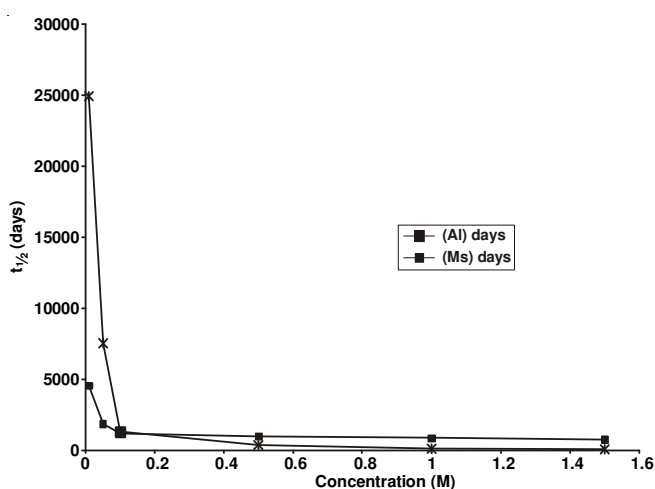

Fig. 15. Plot of half life for mild steel and aluminium against varied HCl concentration in 1 g acetone extract of *Anarcardium occidentale* concentration at 28 ℃
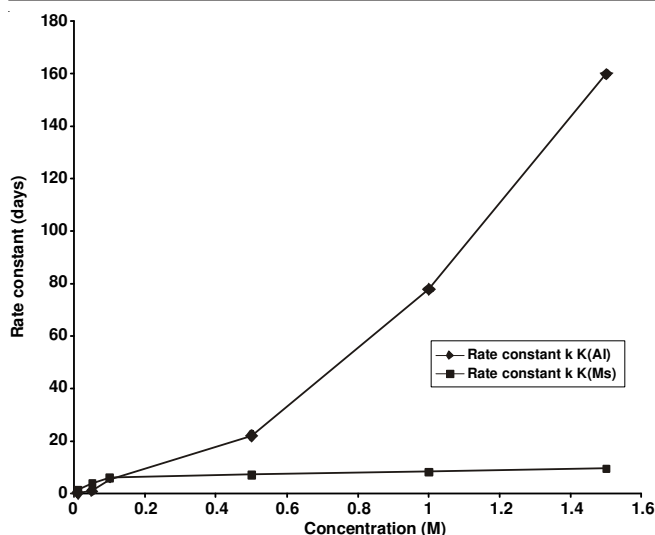
Fig. 16. Plot of rate constant k, for mild steel and aluminium against HCl concentration in 1 g acetone extract of *Anarcardium occidentale* at 28 ℃

where f(θ, x) is the configurational factor, θ the surface coverage, C, the inhibitor concentration in the electrolyte, x, the size factor ratio, a, the molecular interaction parameter and K the equilibrium constant of the adsorption process. The experimental data for mild steel and aluminium was applied to various adsorption isotherm models. It was found that the experimental data fit the Temkin and Freundlich adsorption isotherm given by:

$$\theta = K \ln C \text{ (Temkin)} \tag{8}$$

$$\log \theta = \log K + n \log C \text{ (Freundlich)} \tag{9}$$

Fig. 17 shows the Tenkim plot of θ against log C with varied acetone extract of *Anarcardium occidentale* concentration in (1 M HCl) at 28 ℃.

Linear plots were obtained from the different systems studied at 28 ℃ indicating that the experimental data fit into the Temkin isotherm. The calculated values of the equilibrium constant of adsorption K obtained from the slope of the Temkin plot are shown in Table-7.

From the calculated K values, it was found that the K value for aluminium (0.72) is greater than that of mild steel (0.39) at 28 ℃, showing greater inhibition efficiency for aluminium.

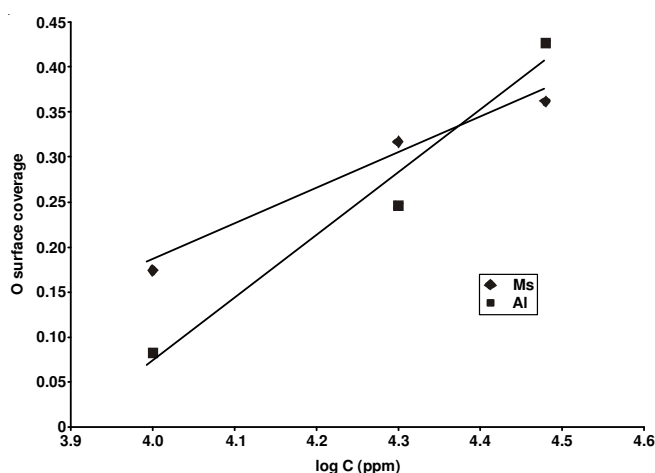Fig. 17. Temkin adsorption isotherm

TABLE-7
CALCULATED VALUES OF FREE ENERGY AND EQUILIBRIUM COEFFICIENT OF ADSORPTION OF ALUMINIUM AND MILD STEEL

Metal coupons	ΔG°_{ads} (KJ mol^{-1})	K
Aluminium	-9.230	0.720
Mild steel	-7.695	0.390

The thermodynamic parameter for adsorption was calculated using the values of K according to the following equation:

$$\Delta G^{\circ}_{ads} = -RT \ln (K \times 55.5) \tag{10}$$

$$\Delta G^{\#} = \Delta H^{\#} - T\Delta S^{\#} = -RT \ln (K^{\#} \times 55.5) \tag{11}$$

The calculated values of ΔG°_{ads} were negative (- 9.23 KJ mol^{-1} for aluminuim and -7.695 KJ mol^{-1} for mild steel) indicating that the adsorption of acetone extract of *Anarcardium occidentale* is spontaneous and that the mechanism of adsorption is physical adsorption (ΔG°_{ads} < 40 KJ mol^{-1}).

Generally, values of (Gibb's free energy or adsorption) ΔG°_{ads} up to -20 KJ mol^{-1} are consistent with electrostatic interaction between charged molecules and a charged metal (which indicates physical adsorption) while those more negative than -40 KJ mol^{-1} involves charge sharing or transfer from inhibitor molecules to the metal surface to form a co-ordinate type of bond (which indicates chemisorption)[9,10].

Conclusion

From the experimental results obtained in the present study, it is undoubtedly true that tannin of *Anacardium occidentale* (cashew nut testa) acts as an acidic corrosion inhibitor for both mild steel and aluminium. The corrosion rate increases with increase in the corrodent (HCl) concentration for both metals and the reverse is true. The inhibitor efficiency (IE %) increases with increased concentration of acetone extract of *Anacardium occidentale*. The tannin exerts its inhibitive behaviours by being adsorbed spontaneously on the surface of the metal surface. The ΔG°_{ads} values are negative and less than - 40 KJ mol^{-1}. Furthermore, the adsorption characteristics of the extract obeys physical adsorption mechanism and follows Temkin and Freundlich adsorption isotherms. The kinetic treatment of the experimental data showed a linear variation which confirms a first order kinetic in the presence and absence of the inhibitor.

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