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Corrosion Inhibition of Copper in Hydrochloric and Tetraoxosulphate (Vi) Acid Solutions Using Aloe Vera Barbadensis Gel

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Authors' contributions

This work was carried out in collaboration between both authors. Author AOJ designed the study, performed the statistical analysis, wrote the protocol, and wrote the first draft of the manuscript and managed literature searches. Author KH managed the analyses of the study and literature searches. Both authors read and approved the final manuscript.

Original Research Article

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ABSTRACT

The inhibition of corrosion of copper in hydrochloric acid and tetraoxosulphate (VI) acid Solutions by Aloe Vera gel has been evaluated by the weight loss method. Inhibition efficiency increased with increase in concentration of the inhibitor system. As the temperature increased the inhibition efficiency decreased. The process follows first order kinetics, and obeys Langmuir adsorption Isotherm. The composition of the active principle of Aloe Vera gel is given.

Keywords: Aloe Vera; Inhibition; copper; weight loss; corrosion.

1. INTRODUCTION

Metals and its alloys are exposed to the action of acids in industrial processes which causes severe problems such as decrease in mass and corrosion of surfaces resulting in economic losses [1]. Copper is one of the most essential metals owing to its large industrial applications. About 65% of copper that is produced is used for electrical applications. The important uses of copper include power generation and transmission of electricity. It is used

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in transformers, motors, bush bars, generators, etc., to provide electricity throughout the country, safely and efficiently [2]. It is also used in wiring, mobile phones and in circuits [3]. It is generally a relatively noble metal; however, it is susceptible to corrosion by acids and strong alkaline solutions, especially in the presence of oxygen or oxidants [4]. Copper is considered to be one of the most important metals which are frequently used in different industrial applications [5]. Copper and its alloys are broadly used in heating systems and condensers. However, these systems should be regularly cleaned due to inlays of carbonates and oxides. Dilute acids are normally used to clean these surfaces [6].

In order to reduce the corrosion of copper metal, several techniques have been applied. The use of inhibitors during acid pickling procedure is one of the most practical methods for protection against corrosion in acid media [6].

By definition, a corrosion inhibitor is a chemical substance that, when added in small concentration to an environment, effectively decreases the corrosion rate [7].The use of inhibitors is one of the best options of protecting metals against corrosion [8]. Corrosion inhibition is a surface process which involves the adsorption of the organic compounds on the metal surface [9].

The applicability of organic compounds as corrosion inhibitors for metals in acidic media has been recognized for a long time [10].The existing data from their research revealed that most organic inhibitors act by adsorption on the metal surface [11]. The adsorption of inhibitors occurs through heteroatoms such as nitrogen, phosphorus, sulphur, triple bonds or aromatic rings. These compounds which are adsorbed on the metallic surface block active corrosion sites [12].

Naturally occurring substances as inhibitors of acid cleaning processes has continued to receive attention as replacement for synthetic organic inhibitors which are sometimes toxic to the environment [10,13,14]. The greatly expanded interest on naturally occurring substances is attributed to the fact that they are cheap, readily available, renewable and ecologically friendly [15]. Literature has shown that that plant materials such as Paradisi juice extract [16], aqueous extract of fenugreek leaves [17], and the root of ginseng, [13] have been used to inhibit the corrosion of various metals. In a similar study, [18] studied the Ethanol Extract of Phyllanthus Amarusas a green inhibitorfor the corrosion of mild steel in H_2SO_4 by weight loss method, hydrogen evolution and IR methods and was found to be a good adsorption inhibitor.

Aloe barbadensis, one of 450 species of Aloe Vera is a succulent perennial herb which grows up to 160 cm tall, without stem or with a short stem up to 30cm long, freely suckering and forming dense groups. Aloe Vera is an important medicinal plant which belongs to the family of Liliacea [19].

The gel, which constitutes the bulk of the leaf substance, serves as the water storage organ for the plant. This gel, which may be removed as a semi-solid “fillet” before processing, contains more than 200 different substances (as shown in Table 1). Chief among these are polysaccharides, glycoproteins, vitamins, mineral and enzymes [20].

Table 1. Phytochemical components of Aloe Vera gel

Class	Compounds
Anthraquinones/anthrones	Aloe-emodin, aloetic-acid, anthranol, aloin A and B (or collectively known as barbaloin), isobarbaloin, emodin, ester of cinnamic acid
Carbohydrates	Pure mannan, acetylated mannan, acetylated glucomannan, glucogalactomannan, galactan, galactogalacturan, arabinogalactan, galatoglucoarabinomannan, pectic substance, xylan, cellulose
Chromones	8-*C*-glucosyl-(2'-*O*-cinnamoyl)-7-*O*-methylaloediol A, 8-*C*-glucosyl-(*S*)-aloesol, 8-*C*-glucosyl-7-*O*-methyl-(*S*)-aloesol, 8-*C*-glucosyl-7-*O*-methyl-aloediol, 8-*C*-glucosyl-noreugenin, isoaloeresin D, isorabaichromone, neoaloesin A
Enzymes	Alkaline phosphatise, amylase, carboxypeptidase, catalase, cyclooxidase, cyclooxygenase, lipase, oxidase, phosphoenolpyruvate carboxylase, superoxide dismutase
Inorganic Compounds	Calcium, chlorine, chromium, copper, iron, magnesium, manganese, potassium, phosphorous, sodium, zinc
Miscellaneous including organic compounds and lipids	Arachidonic acid, γ-linolenic acid, steroids (campestrol, cholesterol, β-sitosterol), triglicendes, triterpenoid, gibberillin, lignins, potassium sorbate, salicylic acid, uric acid
Non-essential and essential amino acids	Alanine, arginine, aspartic acid, glutamic acid, glycine, histidine, hydroxyproline, isoleucine, leucine, lysine, methionine, phenylalanine, proline, threopnine, tyrosine, valine
Proteins	Lectines, lectin-like substance
Saccharides	Mannose, glucose, L-rhamnose, aldopentose
Vitamins	B1, B2, B6, C, β-carotene, choline, folic acid, α-tocopherol

Source: [21]

Aloe Vera gel as commonly called is organic in nature and can be used in the production of green corrosion inhibitors. It is one of the natural inhibitors which has inhibition action on the corrosion of metals. Aloe Vera gel is colorless mucilagenious gel obtained from the parenchymaous cell in fresh leaves of Aloe Vera. It contains various active compounds such as salicylates, magnesium lactate, acemanan, lupeol, campestrol, sterol, linolenic, aloctin and anthraquinones [22].

This present work was designed as contribution to the growing interest on environmentally benign corrosion inhibitors to assess the corrosion inhibitive properties of Aloe Vera gel in hydrochloric and sulphuric acid solutions on Copper at 303K- 353Kand to carry out Kinetic studies of the reaction.

2. EXPERIMENTAL

2.1 Material Preparation

Copper sheets used for this work were bought from Steel village Market, Port Harcourt .The chemical composition of the copper metal are Cu 95.0% and Zn 5.0%. The copper sheets were cut into rectangular specimens of dimension 4cm by 2cm by 0.1cm containing a small hole of about 2mm diameter near the upper edge to allow passage of thread for suspension of the copper coupons into the test solution. The coupon were degreased in absolute ethanol, rinsed with double distilled water and dried in acetone .The treated coupons were then stored over calcium chloride in moisture free desiccators before use for corrosion studies to prevent contamination.

2.2 Preparation of Aloe Vera Gel

The procedure for the preparation of the Aloe Vera gel is similar to that reported by Abiola and James, 2009. Stock solutions of the Aloe Vera gel was prepared by cutting open the Aloe leaves with a clean cutting material to expose the gel content of the leaf.350ml of the gel was squeezed out from 697.9g of the Aloe leaves. The gel was then allowed to filter through a clean white handkerchief in order to get the pure liquid. From the stock solution; the Aloe gel test solutions were prepared at concentrations of 10, 5, 4, 3, 2 and 1 %v/v for solutions of HCl and H_2SO_4 respectively. Thus the highest concentration of 10%v/v was prepared by diluting 10ml of Aloe gel extract with 90ml of HCl or H_2SO_4. Other concentrations (5%, 4%, 3%, 2% and 1%.vol/vol) were prepared by diluting 5ml, 4ml, 3ml, 2ml and 1ml of Aloe gel extract with 95ml, 96ml, 97ml, 98ml and 99ml of HCl or H_2SO_4 respectively).

2.3 Weight Loss Method

The copper metal, sheet of 0.1cm thickness and 98.79% purity used for weight loss measurement were of dimension 4cm by 2cm. The total geometric surface area of the coupons exposed to the corrodent was 16.00cm^2 (since both faces of the coupons were immersed in the corrodent). The average weight of the copper coupons was 4.48 - 4.58g.

Fifteen 250ml beakers, which separately contained 100ml of 10%, 5%, 4%, 3%, 2% 1% v/v and 2M of HCl and H_2SO_4 which served as the control respectively were maintained at 303K, 313K, 323K, 333K and 353K constituting two set of experiments for each acid. Previously weighed copper coupons were each suspended in each beaker through a 0.1cm hole in diameter.

The copper coupons in HCl and H_2SO_4 acid solutions at 303K, 313K, 323K, 333K and 353K were retrieved at 24hours interval progressively for 168 hours (7 days) respectively.

The copper coupons retrieved were immediately washed thoroughly in water, rinsed in acetone and dried in an oven to constant weight before final weighing.

The coupons retrieved were immersed in a solution of 20% sodium hydroxide to terminate the corrosion reaction. The coupons was scrubbed with brittle brush several times inside water to remove corrosion products, dried in ethanol and acetone and then weighed.

The weight loss was calculated in grams as the difference between the initial weight prior to immersion, and weight after the removal from acid solution.

Triplicate experiments were performed in each case and the mean values reported to the nearest 0.0001g on an AB54 AR analytical weighing balance.

The inhibition efficiency (%I) was computed using equation 2.1:

i. Inhibition efficiency at 303K, 313K, 323K, 333K and 353K.

$$\%\text{Inhibition Efficiency} = \frac{\Delta W_B - \Delta W_1}{\Delta W_B} \times \frac{100.}{1} \quad \ldots\ldots\ldots\ldots\ldots\ldots (2.1)$$

Where ΔW_B and ΔW_1 are the weight loss of copper metal coupons without and with inhibitor respectively.

The corrosion rate (W) was computed using the equation 2.2:

ii. Corrosion rate at 303K, 313K, 323K, 333K and 353K.

a) $$\text{Corrosion rate } (g/cm^2/day) = \frac{\Delta W}{\text{Area } (cm^2) \times \text{day}} \quad \ldots\ldots\ldots\ldots\ldots\ldots (2.2)$$

Where ΔW is the change in weight or weight loss (g), A is the area of the specimen (cm^2) and t is the exposure time in days.

3. RESULTS AND DISCUSSION

3.1 Effect of Corrodent Concentration on Copper Corrosion

Fig. 3.1 and Fig. 3.2 shows that copper corrodes in different concentrations of HCl and H_2S0_4 respectively, since there was a decrease in original weight of copper. The corrosion is attributed to the presence of water, air and H^+, which accelerates the corrosion process. The corrosion of copper in HCl and H_2S0_4 increases with the concentration of the acid and time. Similar results were obtained at 313K, 323K, 33K and 353K. This observation is attributed to the fact that the rate of chemical reaction increases with increasing concentration [23].

3.2 Effect of Temperature

The electrochemical nature of corrosion causes the corrosion rate to increase exponentially with rise in temperature (as seen in Figs. 3.5 and 3.6), according to Arrhenius-type dependence. Thus the influence of temperature on the corrosion behavior of Cu in 2M HCl and 2M H_2SO_4 in the absence and presence of Aloe gel of varying concentrations were investigated by weight loss method at 300K, 313K.323K, 333K and 353K. In examining the effect of temperature on the corrosion inhibition process, the apparent activation energies (E_a) were calculated from the Arrhenius equation [7].

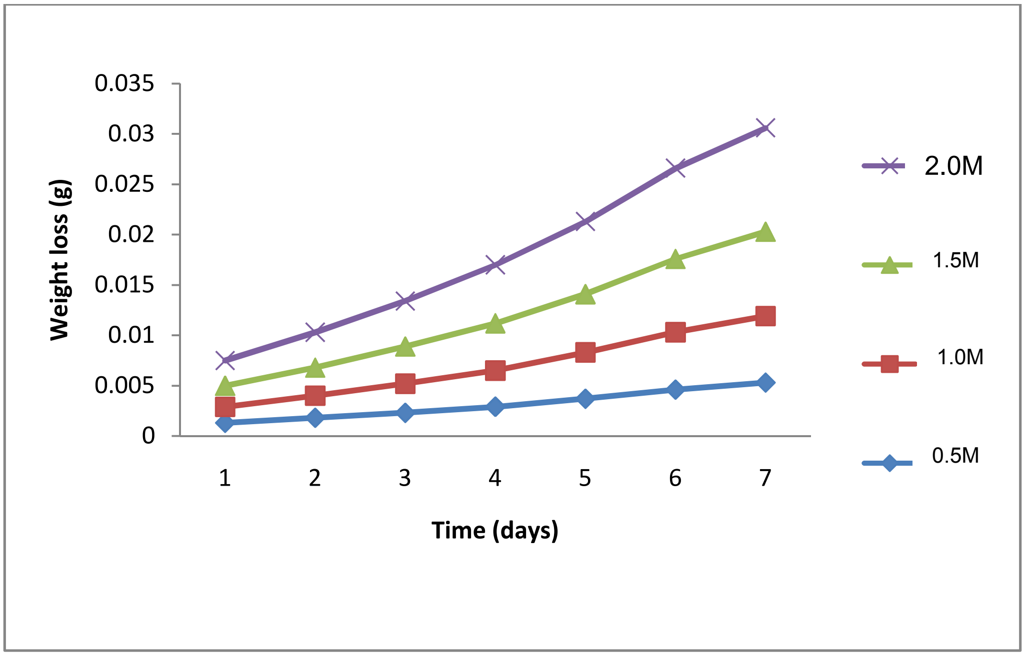

Fig. 3.1. Variation of weight loss (g) with time (days) for different concentrations of HCl solution at 303K

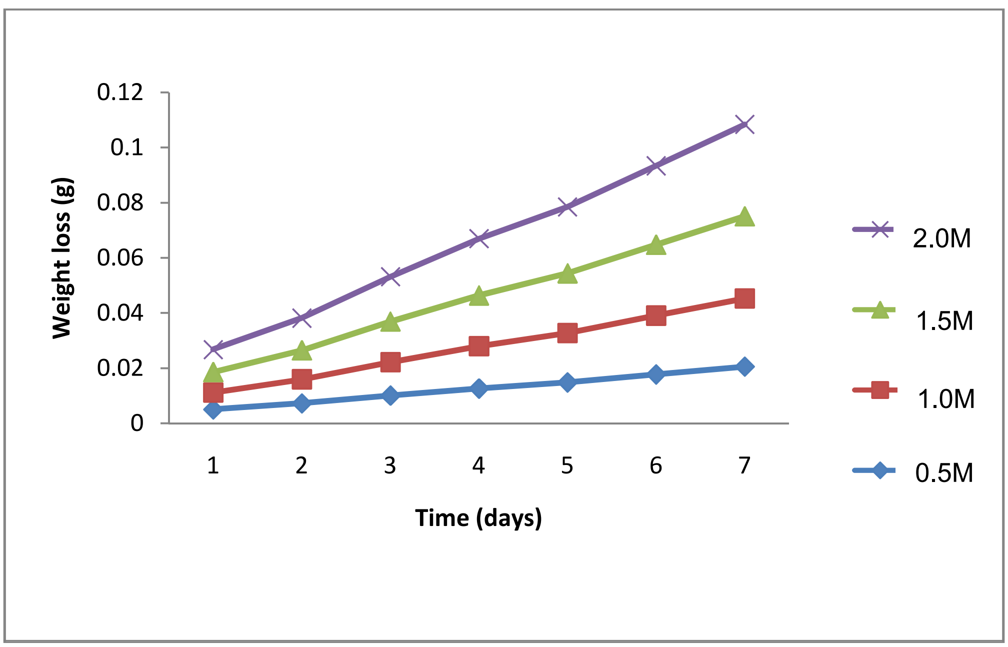

Fig. 3.2. Variation of weight loss (g) with time (days) for different concentrations of H_2S0_4 solution at 303K

$$Ea = \frac{2.303RT_1T_2\,(\log k_2/k_1)}{T_2 - T1} = \frac{Jmol^{-1}}{1000} = KJmol^{-1} \quad \ldots\ldots\ldots\ldots\ldots (2.3)$$

Where

R = is the gas constant in Joule (J)
T = Absolute temperature in Kelvin
R = 8.314 $Jmol^{-1}\ K^{-1}$

K_1 and K_2 are the rate constants at T_1 (300K) and T_2 (313K) respectively or T_1 (313K) and T_2 (323K) and so on for the rest of the temperatures.

The Activation energies were calculated from equation 2.3 and recorded in Table 4. The values of Ea ranged from 65.88 to 31.60 KJ/mol and 51.21 to 15.08 KJ/mol for HCl and H_2SO_4 respectively. These values are less than the threshold value of 80 KJ/mol required for chemical adsorption [18], hence the adsorption of Aloe Vera gel on the surface of copper supports the mechanism of physical adsorption.

3.3 The Inhibitive Effect of Aloe Vera Gel

The values of percentage inhibition efficiency and corrosion rate obtained from weight loss method at different concentrations of the Aloe gel at 300K-353K are summarized in Tables 2 and 3 above. Inspection of the data in the tables above reveals that the addition of Aloe gel decreases the corrosion rate of copper in HCl and H_2S0_4 acidic solutions. From Figs. 3.3 and 3.4 below it is also observed that the inhibition efficiency increases as the concentration of the Aloe gel is increased.

3.4 The Active Component Responsible for the Inhibitory Action of Aloe Vera Gel

The observed inhibition of the Aloe gel could be attributed to the adsorption of the components on the copper coupon surface. The formed layer, of the adsorbed molecules, isolates the metal surface from the aggressive medium leading to decreasing of the corrosion rate. The chemical components of the exudate of Aloe Vera was studied and reported in literature [22]. As earlier stated in the introductory remarks, Aloe Vera gel is a complex mixture of as salicylates, magnesium lactate, acemanan, lupeol, campestrol, sterol, linolenic, aloctin and anthraquinones. Other components include saponins, glycosides and vitamins, which implies that they contain heteroatoms such as oxygen and nitrogen in its molecule. The corrosion inhibition of copper may be attributed to adsorption of Aloe gel exudate onto the metal surface. Owing to complex chemical composition of the exudate, it is quite difficult to assign the inhibitive effect to a particular constituent. Further investigation using surface analytical techniques will enable the characterization of the active materials in adsorbed layer and assist in identifying the most active ingredients. It is worthy to note that in acidic media, nitrogen atoms accept protons from the acid solution and form cations which electrostatically are attracted to the pre-adsorbed Cl^{-1} or SO_4^{-2} anions on the copper coupon surface [24].

Table 2. Values of corrosion rate ($gdm^{-2}day^{-1}$) and inhibition efficiency (% I) for copper in 2M HCl

Inhibitor concentration (%)	Inhibition efficiency (% I)					Corrosion rate ($gdm^{-2}day^{-1}$)X10^{-4}				
	303K	313K	323K	333K	353K	303K	313K	323K	333K	353K
1.0	37.77	24.71	18.75	14.07	11.40	1.75	4.18	5.68	7.25	8.25
2.0	40.10	28.08	20.53	16.29	13.42	1.68	4.00	5.56	7.06	8.06
3.0	44.44	35.95	33.92	26.66	21.47	1.56	3.56	4.62	6.25	7.31
4.0	53.33	43.82	37.50	32.59	28.85	1.31	3.12	4.37	5.68	6.62
5.0	64.44	55.05	49.10	43.70	37.58	1.00	2.5	3.56	4.75	5.81
10.0	71.11	65.16	60.71	53.33	48.99	0.812	1.93	2.75	3.93	4.75

Table 3. Values of corrosion rate ($gdm^{-2}day^{-1}$) and inhibition efficiency (% I) for copper in 2M H_2SO_4

Inhibitor concentration (%)	Inhibition efficiency (% I)					Corrosion rate ($gdm^{-2}day^{-1}$)X10^{-4}				
	303K	313K	323K	333K	353K	303K	313K	323K	333K	353K
1.0	30.66	21.77	16.29	10.95	7.59	3.25	6.06	7.06	8.12	9.12
2.0	34.66	25.01	18.51	13.69	10.75	3.06	5.81	6.87	7.87	8.81
3.0	37.33	33.87	28.14	21.23	17.72	2.93	5.12	6.06	7.18	8.12
4.0	45.33	40.32	35.55	27.37	25.94	2.56	4.26	5.43	6.62	7.31
5.0	56.00	50.8	45.92	39.04	34.81	2.06	3.81	4.56	5.56	6.43
10.0	64.10	58.87	54.07	47.94	46.20	1.68	3.18	3.87	4.75	5.31

Table 4. Activation energies of Aloe Vera gel exudate on the surface of copper for HCl and H_2SO_4

Activation energy $Kjmol^{-1}$ HCl		Activation energy $kJmol^{-1}$ H_2SO_4 average activation energy $kJmol^{-1}$	
303K-313K	323K-313K	303K-313K	323K-313K303K-313K 323K-313K
60.71	30.43	57.72	11.83
66.25	33.88	56.51	13.01
63.04	24.67	46.44	17.4565.8851.2131.6015.08
69.18	30.79	48.61	16.35
67.75	37.02	49.29	14.13
68.35	32.85	48.72	17.72

The following components with structures contain electron rich oxygen and nitrogen that could serve as good active ingredients which are responsible for corrosion inhibition in Aloe Vera gel.

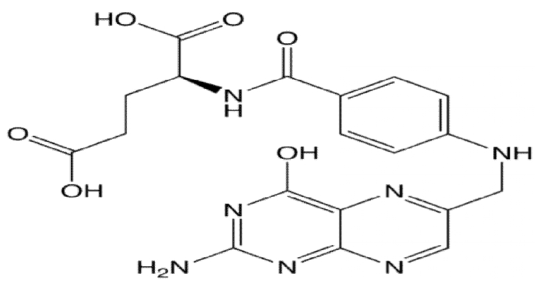

Fig. 3a. Folic Acid (A Vitamin)

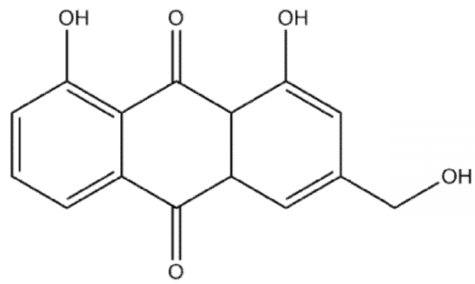

Fig. 3b. Aloe Emodin (C15H10O5) (An Anthraquinone)

3.5 Effect of Concentration Increase on the Inhibtion Efficiency of Aloe Vera Gel Exudate on the Surface of Copper for HCl and H_2so_4

From Table 2, it was observed that the percentage inhibition efficiency increases with the concentration of the inhibitor and temperature. This observation is supported by Figs. 3.3 and 3.4.

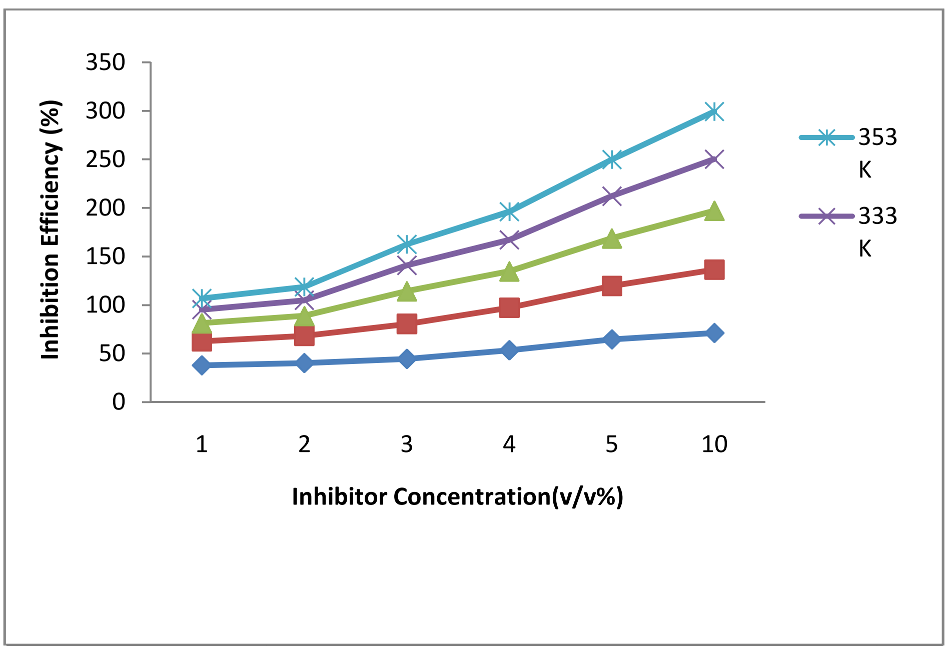

Fig. 3.3. Average inhibition efficiency (%) for the inhibition of copper in 2M HCl solution by Aloe Vera gel at different temperatures

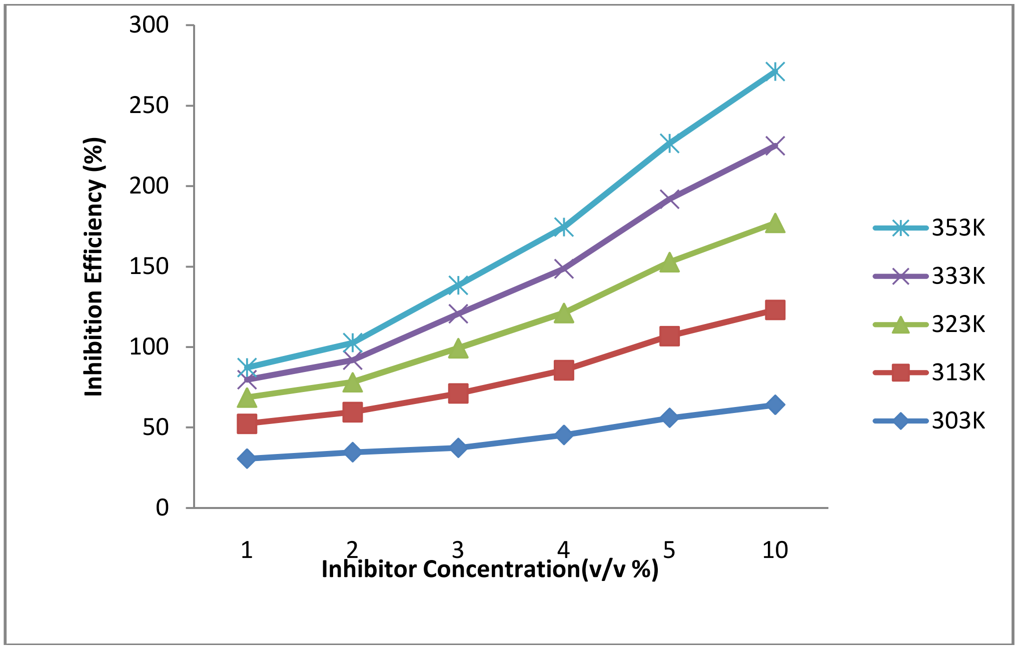

Fig. 3.4. Average inhibition efficiency (%) for the inhibition of copper in 2M H_2SO_4, solution by Aloe Vera gel at different temperatures

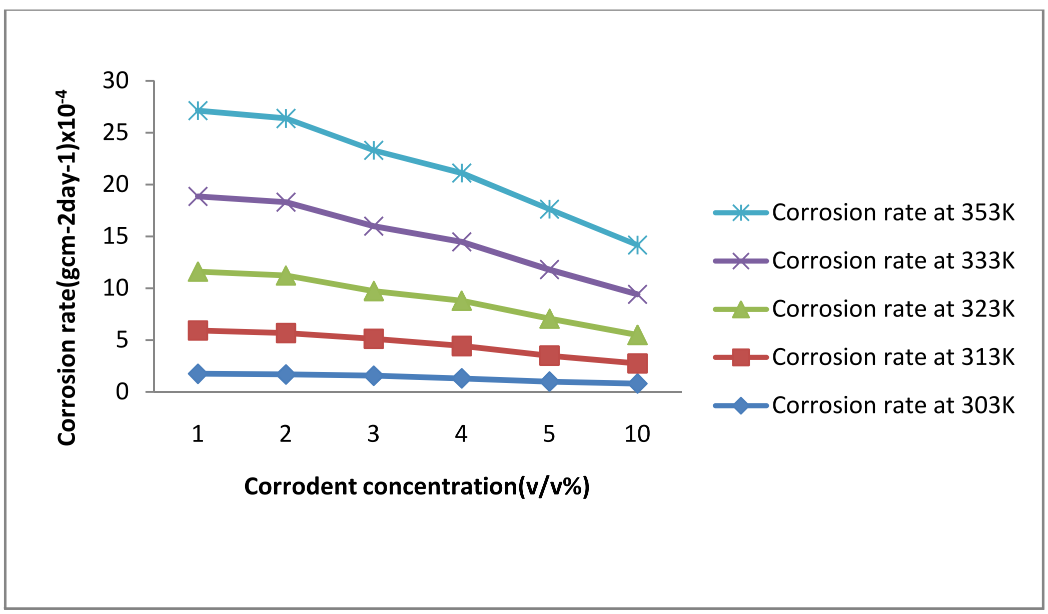

Fig. 3.5. Variation of weight loss (g) of copper with time (days) for different concentrations of HCl solutions at different temperatures

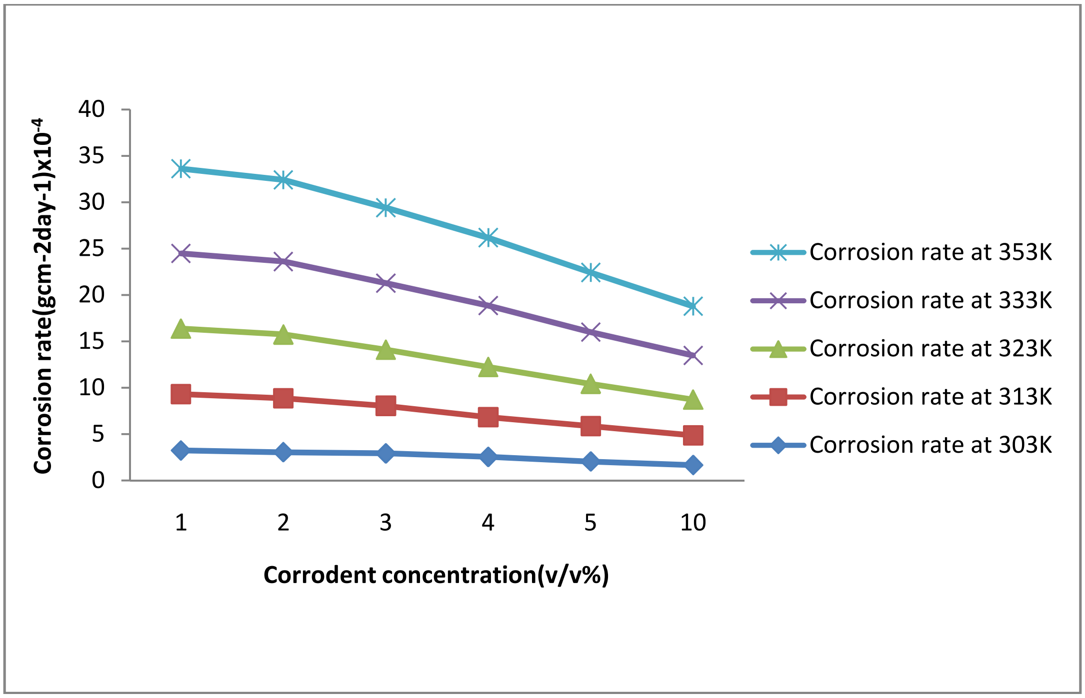

Fig. 3.6. Variation of weight loss (g) of copper with time (days) for different concentrations of H_2SO_4 solutions at different temperatures

3.5 Adsorption Consideration

The effectiveness of organic compounds as corrosion inhibitors can be ascribed to the adsorption of molecules of the inhibitors through their polar functions on the metal surface. Surface coverage values were evaluated from the weight loss measurements assuming direct relationship between inhibition efficiency and surface coverage as follows: % I = θ x 100. The surface coverage values were fitted to different adsorption isotherm models and best results fitted the Langmuir isotherm.

Langmuir isotherm is given by the expression:

$$\frac{C_{inh}}{\theta} = \frac{1}{K_{e.c.a}} + C_{inh} \qquad (2.4)$$

Where θ is the surface coverage, C_{inh} is the concentration, $K_{e.c.a}$ is the equilibrium constant of the adsorption process. The plot of C/ θ against C is shown in Figs. 3.7 and 3.8.

The plot at the various temperatures studied which gives a straight line fit the adsorption of Aloe Vera gel on the copper metal to Langmuir adsorption isotherm.

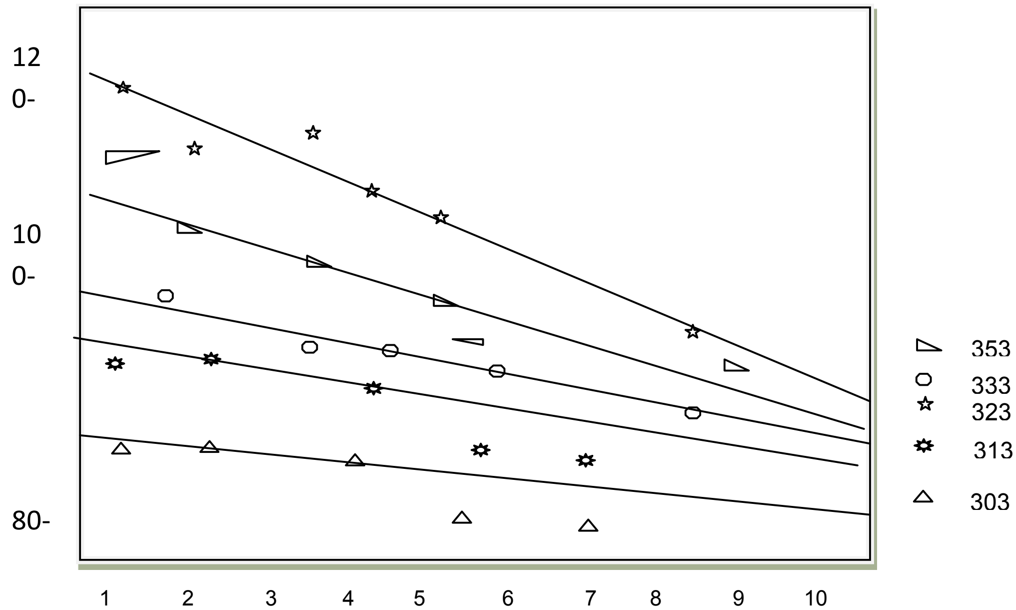

Fig. 3.7. Langmuir adsorption isotherm (plotted as C/θ) versus C for the inhibition of copper in 2MHCl solution by Aloe Vera gel

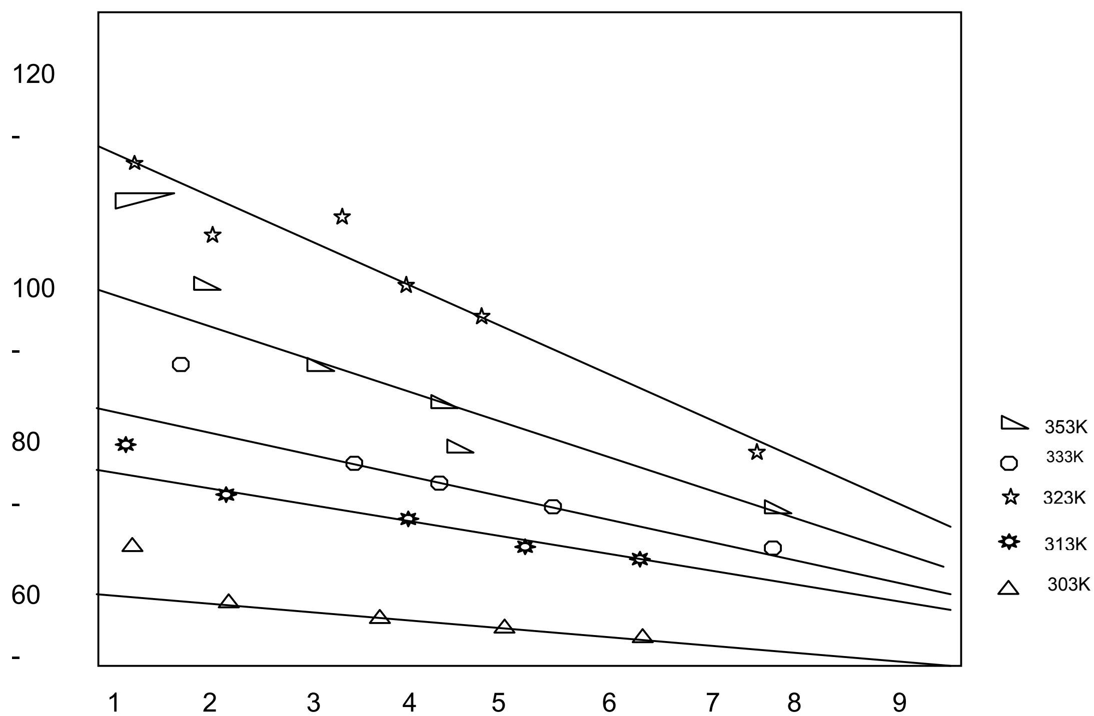

Fig. 3.8. Langmuir adsorption isotherm (plotted as C/θ) versus C for the inhibition of copper in 2MH_2SO_4 solution by Aloe Vera gel

4. CONCLUSION

From the results obtained from this study, the following conclusions have been made:

(i) The rates of corrosion of copper in HCl and H_2SO_4 acid solutions without inhibitor increases with increase in acid concentration and temperature range studied.
(ii) The methods used show that Aloe Vera gel inhibited the corrosion of copper to a reasonable extent in both HCl and H_2SO_4 acid solutions.
(iii) The additive used in this investigation (Aloe Vera gel) inhibited the acid corrosion of copper in HCl and H_2SO_4 to different degrees.
(iv) The percentage inhibition efficiency (%) of the inhibitors is in the other of HCl> H_2SO_4, that is there was better corrosion inhibition in HCl than H_2SO_4.
(v) Kinetic treatment of the results of the corrosion of copper in HCl and H_2SO_4in both inhibited and unhibited reactions confirm a first order type of mechanism.
(vi) The values of the activation energy obtained in the presence and absence of inhibitors are below 80KJmol^{-1} (see Table 4). The inhibition is probably due to the adsorption of the inhibitor which was physically adsorbed on the surface of the copper metal.
(vii) The adsorption fits well to the Langmuir adsorption Isotherm.
(viii) The results obtained suggest that Aloe Vera gel is a corrosion inhibitor for copper metals in HCl and H_2SO_4 and they can be used to replace toxic and nonbiogradable inhibitors.

COMPETING INTERESTS

Authors declare that there are no competing interests.

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