

Effect of sulphuric acid concentration on the inhibiting action of 0.001M adenine solution during the corrosion of AISI 304L

¹Ofuyekpone O., ²Emordi N. and ²Utu O. G.

¹Department of Metallurgical Engineering, Delta State Polytechnic, Ogwashi-Uku, Delta State, Nigeria

²Department of Welding and Fabrication, Delta State Polytechnic, Ogwashi-Uku, Delta State, Nigeria

ABSTRACT

In this research the effect of sulphuric acid concentration on the inhibiting action of 0.001M adenine solution during the corrosion of AISI 304L has been investigated. The research was achieved by using 0.001M concentration of adenine solution and varied sulphuric acid concentration of 0.1M, 0.3M, 0.5M, 0.7M, 0.9M and 1.0M; readings were obtained every 240hours (10 days) for a period of 1200hours (50 days). Weight loss, corrosion penetration rate, inhibition efficiency and degree of surface coverage were calculated. Results obtained revealed that adenine is an effective and safe corrosion inhibitor for AISI 304L in sulphuric acid solutions within the concentration range of the acid investigated. The inhibition mechanism involves the adsorption of protonated adenine molecules onto the 304L surface. Analysis of the results showed that the inhibition efficiency and surface coverage decreased with increasing sulphuric acid concentration, causing an increase in corrosion penetration rate and weight loss as the concentration of the sulphuric acid was increased. The corrosion penetration rate, however, decreased with increased immersion time. Also, the inhibition efficiency and surface coverage decreased with increasing immersion time.

Keywords: Corrosion, AISI 304L Steel, Inhibition efficiency, Degree of Surface coverage, Sulphuric Acid Solution

INTRODUCTION

Stainless steels are the first 'step-up' from carbon steels in corrosion resistance [1]

Corrosion is the destruction of a material resulting from exposure and interaction with the environment [2]. Corrosion remains one of the most severe limitations for the use of various steels in the chemical and petrochemical industries. Millions of dollars are lost each year because of corrosion. Much of this loss is due to the corrosion of iron and steel [3].

The corrosion of stainless steels in acidic solution is of fundamental academic and industrial concern that has received a considerable amount of attention [4-5]. The most important field of application being acid pickling, industrial acid cleaning, acid descaling and oil well acidizing. Because of the general aggressiveness of acid solutions, inhibitors are commonly used to reduce the corrosion attack on metallic materials. Organic adsorption compounds are effective as corrosion inhibitors for corrosion of different types of stainless steels in acidic medium because of the functional group containing heteroatom such as nitrogen, sulphur and oxygen [6].

It is also pertinent to state that over the years, considerable efforts have been deployed to finding suitable or green (safe and friendly) corrosion inhibitors of organic origin in various corrosive media [7]. Several factors including cost and amount, easy availability and most important safety to environment and its species need to be considered when choosing an inhibitor [8]. The use of inhibitors is one of the most practical methods for protection against corrosion and prevention of unexpected metal dissolution [9]. Treatments with organic compounds, as corrosion inhibitors, are frequently proposed in order to improve anticorrosion protection in acidic media [10].

In general, inhibitors are classified as anodic, cathodic or mixed [11].

In the past two decades, research in the field of “green” corrosion inhibitors has been aimed at using cheap, effective molecules with low or “zero” environmental impact. Among the numerous organic compounds tested and industrially applied as corrosion inhibitors, non-toxic ones are far more strategic now than in the recent past. These compounds include amino acids and its derivatives as mimosa tannin or isatin etc., which have been tested for various metals and alloys [12, 13 - 21].

Adenine is a purine derivative also called Amino-6-purine. Its IUPAC name is 7H-purin- 6-amine. It has a molecular formula of $C_5H_5N_5$ and its structural formula is as shown in Fig. 1

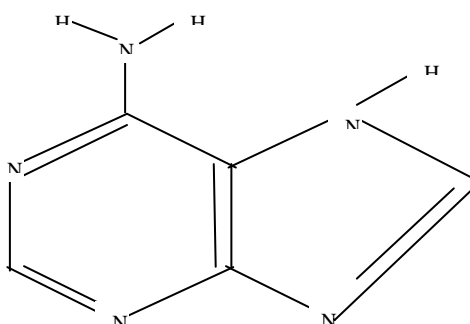


Figure 1. Structural formula of Adenine

The molecular mass of Adenine is 135.13g/mol. It is solid at standard temperature and pressure, and melts at 360°C . It is biodegradable. It is soluble in water and has a density of 0.99172g/cm^3

Tetraoxosulphate (VI) acid (Sulphuric acid) is the corrosive media used in this work. It is produced more than any other chemical in the world. It has large scale uses covering nearly all industries, such as fertilizer industries, petroleum refinery, paint industry, steel pickling, extraction of non-metals, and manufacture of explosives [22].

In this work, varied concentration of the acid is used simultaneously with 0.001M concentration of Adenine solution as an inhibitor. The present investigation aims at evaluating the effect of sulphuric acid concentration on the inhibiting action of 0.001M adenine solution during the corrosion inhibition of AISI 304L

MATERIALS AND METHODS

Materials

Austenitic stainless steel (alloy 304L) rod of 10mm in diameter was used in this research work. The chemical analysis of the alloy 304L (UNS S30403) is shown in Table 1. Adenine is the organic compound used as the inhibitor while tetraoxosulphate VI acid is the corrosive agent used in this research work. The structure of the adenine is shown in Figure 1.

Table 1: Chemical Composition of Investigated Alloy

Elements	C	Si	Mn	P	S	Cr	Ni	Mo	Cu	V	Al	Sn	Ti	Fe
Weights %	0.03	0.42	1.67	0.044	0.033	18.19	7.16	0.29	0.42	0.059	0.013	0.012	0.005	71.85

Experimental Procedure

The austenitic stainless steel rod was prepared to a dimension of 10mm x 10mm. The samples totaled 60. The samples were degreased using acetone by immersing them for a period of about 5minutes, and then washed (rinsed) with double distilled water and dried. The samples were shared into twelve different containers of the ‘solutions’ labeled A_1 , A_2 , $B_1 - F_2$ and containing five (5) samples each. The samples with the subscript 1 are the uninhibited samples at the specified concentrations of the sulphuric acid while the samples with the subscript 2 are the inhibited samples at the specified concentrations of the sulphuric acid. In all cases, a sample was taken from each container at ten days interval, rinsed and dried, and then their corrosion rate determined by weight loss. **Equations (1-4)** were used to obtain the results of **Figures 2-13**.

$$\text{CPR} = \frac{87.6 * W}{\text{DAT}} \quad (1)$$

$$SML = \frac{W}{A} \tag{2}$$

$$IE = \frac{CR_o - CR_i}{CR_o} * 100 \tag{3}$$

$$\Theta = \frac{W_{t_o} - W_{t_i}}{W_{t_o}} \tag{4}$$

Where W = weight loss, D = density. A = area, T = immersion time, CRo = corrosion penetration rate in the absence of inhibitor, CRi = corrosion penetration rate in the presence of inhibitor, Wto = weight loss in the absence of inhibitor, and Wti = weight loss in the presence of inhibitor.

RESULTS

The results of the weight loss, corrosion penetration rate, inhibition efficiency and degree of surface coverage in varied concentration of sulphuric solutions with 0.001M concentration of Adenine as inhibitor, and without adenine addition, during the corrosion of AISI 304L are given in **Figures 2-13**.

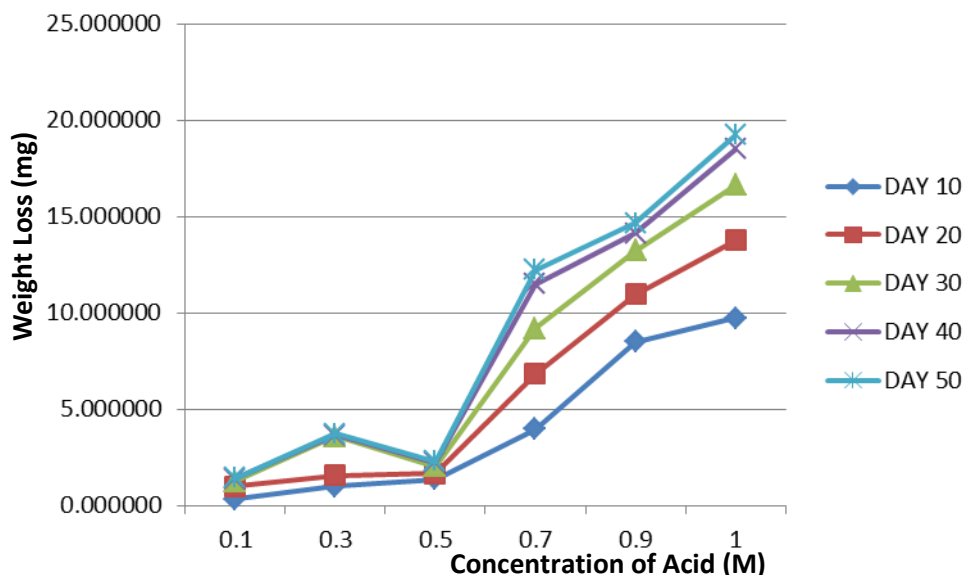


Figure 2: Graph of Weight loss (mg/cm²) Vs Acid conc. (M)

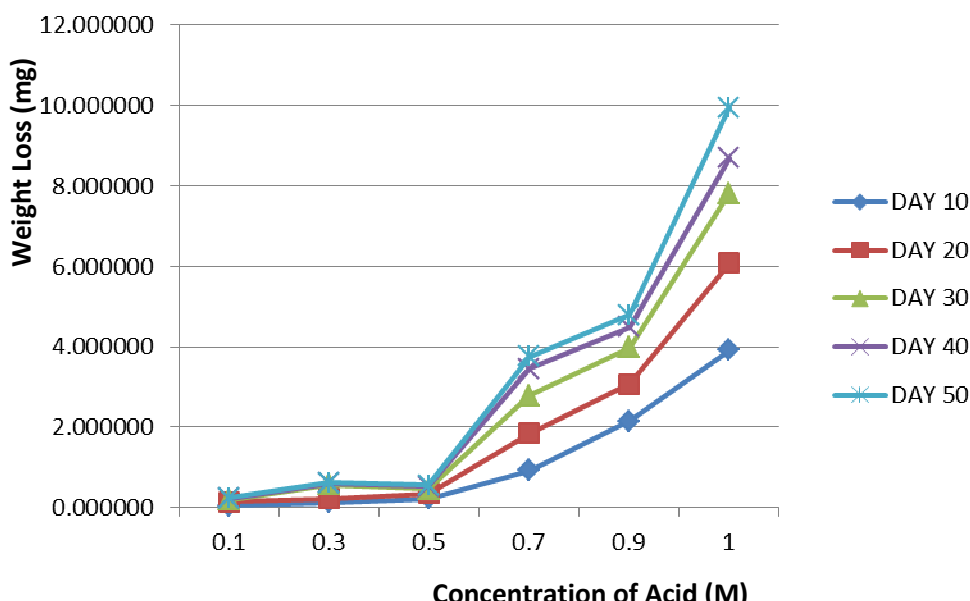


Figure 3: Graph of Weight loss (mg/cm²) Vs Acid conc. (M)

The **Figure 2** shows the variation of the weight loss data (mg/cm^2) with Sulphuric acid concentration (M) recorded for alloy 304L in various concentrations of H_2SO_4 at 10 days interval for 50 days and without Adenine addition

The **Figure 3** shows the variation of the weight loss data (mg/cm^2) with the Sulphuric acid concentration (M) recorded for alloy 304L in 0.001M Adenine solution with various concentrations of H_2SO_4 at 10 days interval for 50 days.

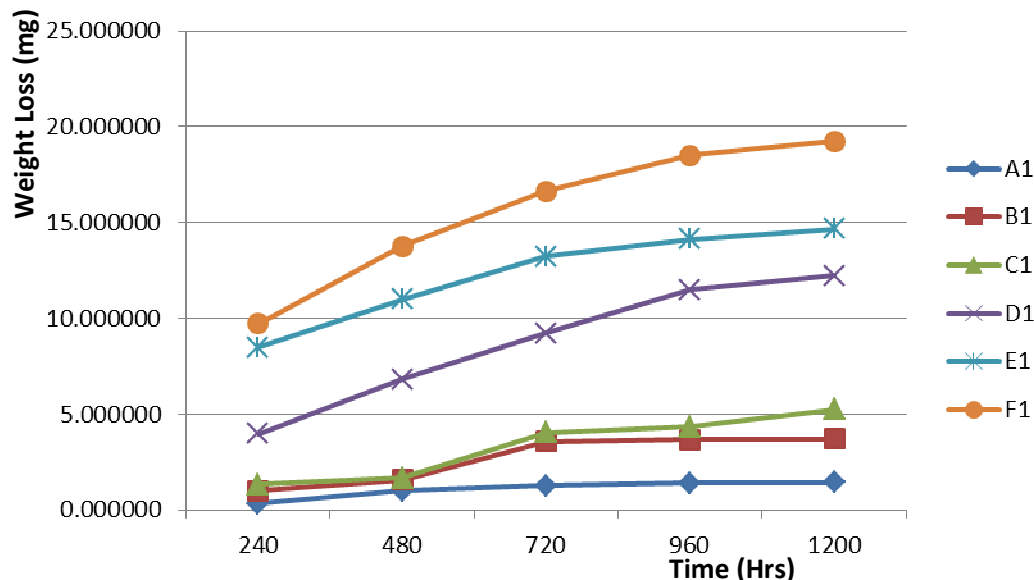


Figure 4: Graph of weight loss (mg/cm^2) Vs Time (hrs)

The **Figure 4** shows the variation of the weight loss data (mg/cm^2) with the immersion time (hrs) recorded for alloy 304L in various H_2SO_4 concentrations without inhibitor (adenine) added. (A1) 0.1M H_2SO_4 ; (B1) 0.3M H_2SO_4 ; (C1) 0.5M H_2SO_4 ; (D1) 0.7M H_2SO_4 ; (E1) 0.9M H_2SO_4 ; (F1) 1.0M H_2SO_4

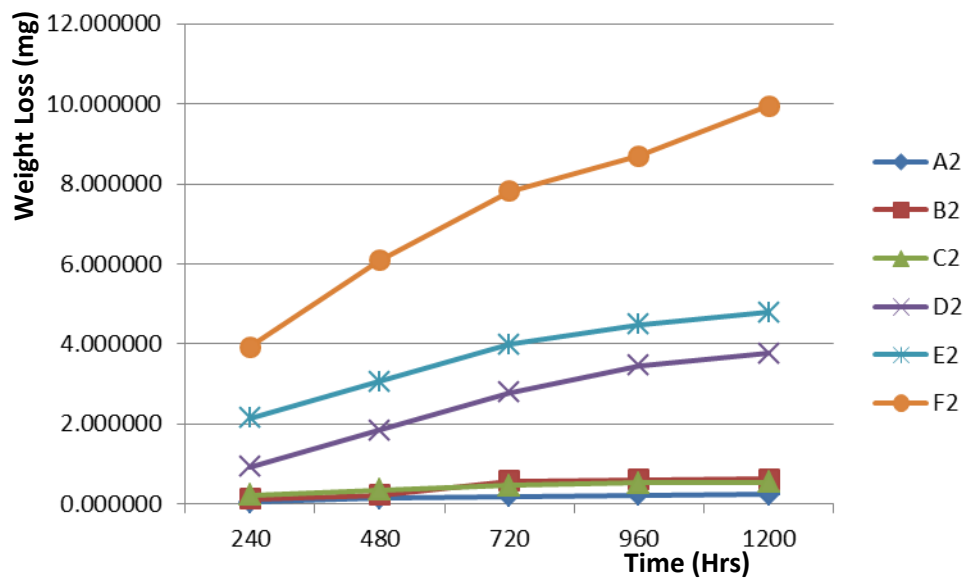


Figure 5 : Graph of weight loss (mg/cm^2) Vs Time (hrs)

The **Figure 5** shows the variation of the weight loss data (mg/cm^2) with the immersion time (hrs) recorded for alloy 304L in 0.001M Adenine solution with various concentrations of H_2SO_4 . (A₂) 0.1M H_2SO_4 ; (B₂) 0.3M H_2SO_4 ; (C₂) 0.5M H_2SO_4 ; (D₂) 0.7M H_2SO_4 ; (E₂) 0.9M H_2SO_4 ; (F₂) 1.0M H_2SO_4

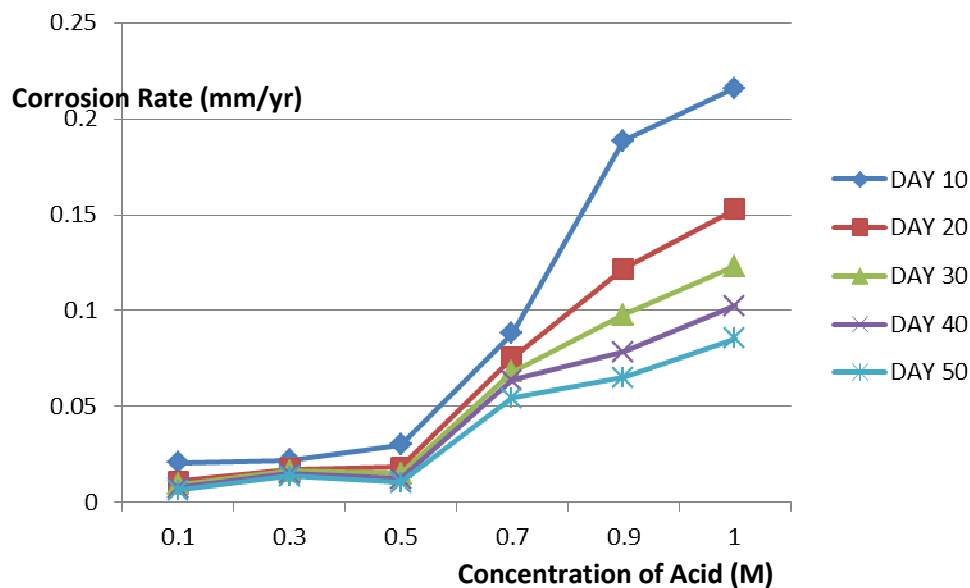


Figure 6 . Graph of Corr. Rate (mm/yr) Vs H₂SO₄ Conc. (M)

The **Figure 6** shows the variation of the Corrosion penetration Rate (mm/yr) against the Sulphuric acid Concentration (M) recorded for alloy 304L in various concentrations of H₂SO₄ at 10 days interval for 50days and without Adenine addition.

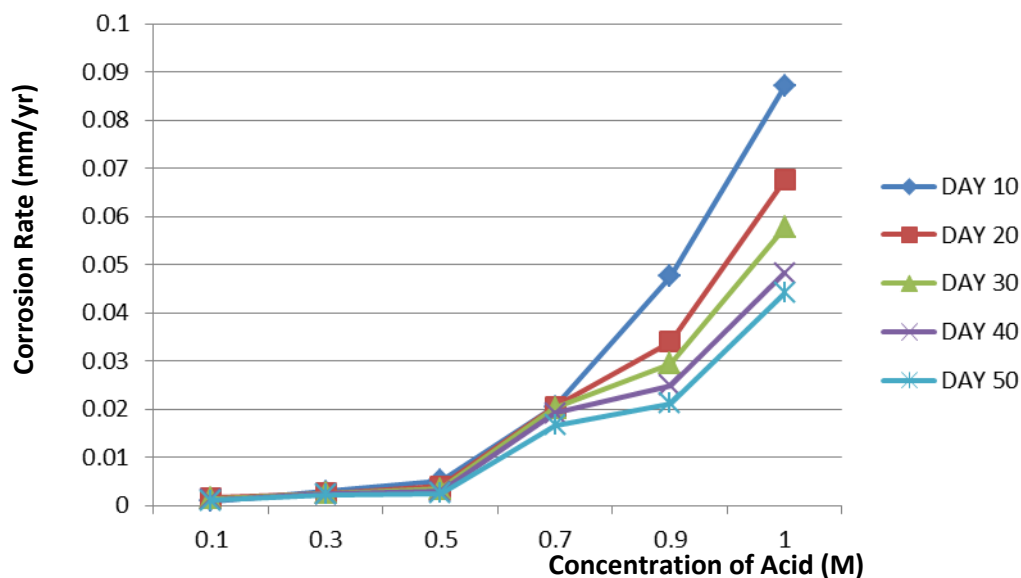


Figure 7 . Graph of Corr. Rate (mm/yr) Vs H₂SO₄ Conc. (M)

The **Figure 7** shows the variation of the Corrosion penetration Rate (mm/yr) against the Sulphuric acid Concentration (M) recorded for alloy 304L in 0.001M Adenine solution as inhibitor at 10 days interval for 50days

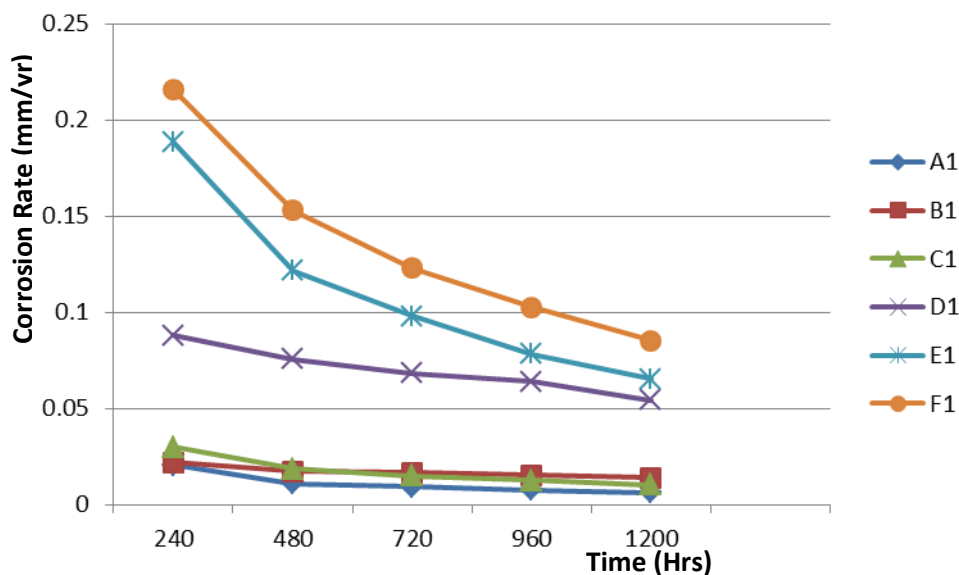


Figure 8 : Graph of Corrosion Rate(mm/yr) Vs (Time hrs)

The **Figure 8** shows the variation of the Corrosion penetration rate (mm/yr) with the immersion time (hrs) recorded for alloy 304L in various concentrations of H₂SO₄ solution without inhibitor (Adenine) added. (A₁) 0.1M H₂SO₄; (B₁) 0.3M H₂SO₄; (C₁) 0.5M H₂SO₄; (D₁) 0.7M H₂SO₄; (E₁) 0.9M H₂SO₄; (F₁) 1.0M H₂SO₄.

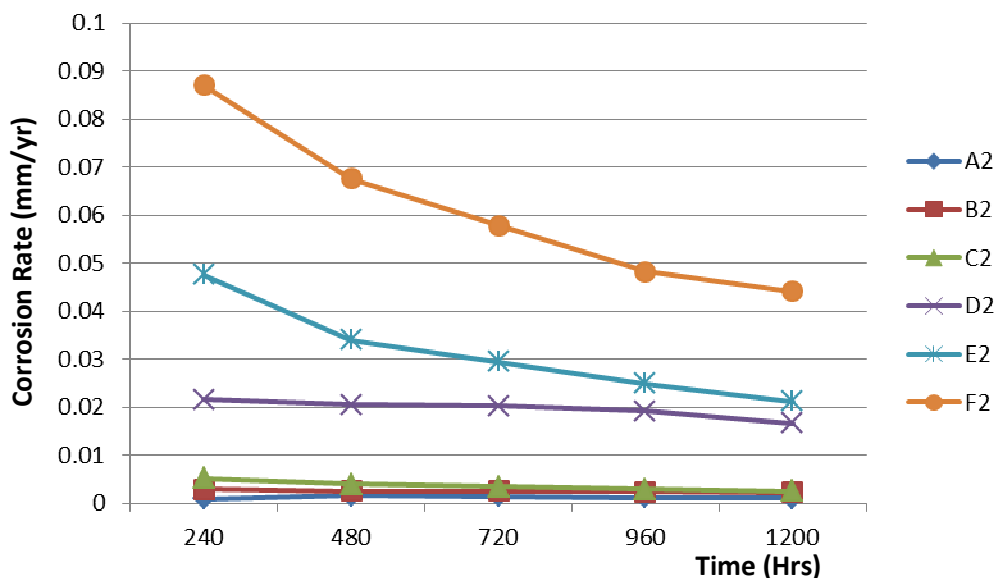


Figure 9 : Graph of Corr.Rate(mm/yr) Vs (Time hrs)

The **Figure 9** shows the variation of the Corrosion penetration rate (mm/yr) with the immersion time (hrs) recorded for alloy 304L in various concentrations of H₂SO₄ solution and 0.001M Adenine as inhibitor. (A₂) 0.1M H₂SO₄; (B₂) 0.3M H₂SO₄; (C₂) 0.5M H₂SO₄; (D₂) 0.7M H₂SO₄; (E₂) 0.9M H₂SO₄; (F₂) 1.0M H₂SO₄.

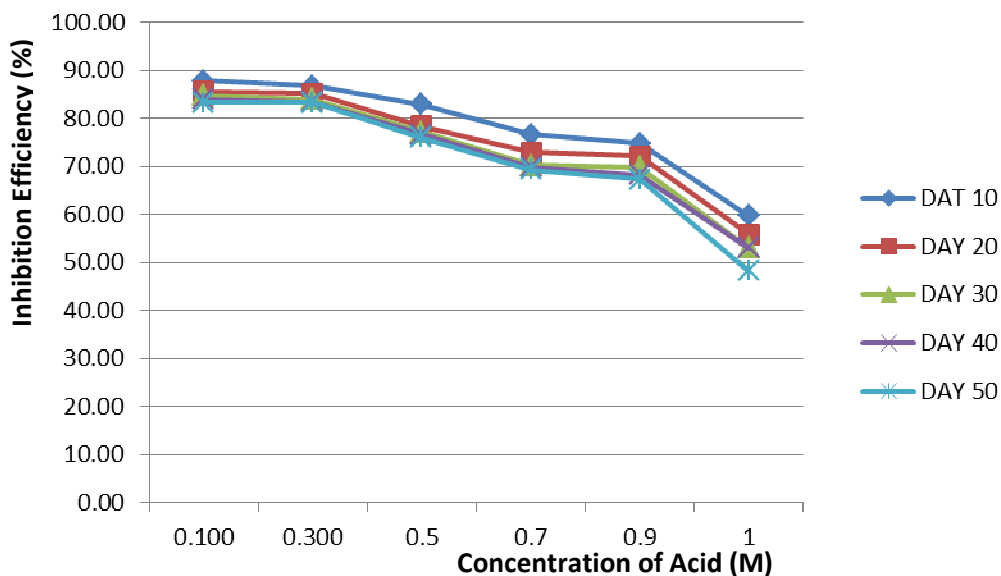


Figure 10: Graph of Inhibition Efficiency (%) Vs Acid Conc. (M)

The **Figure 10** shows the variation of the Inhibition Efficiency (%) with various concentration of the H_2SO_4 recorded for alloy 304L in 0.001M Adeninesolution at 10 days interval for 50days

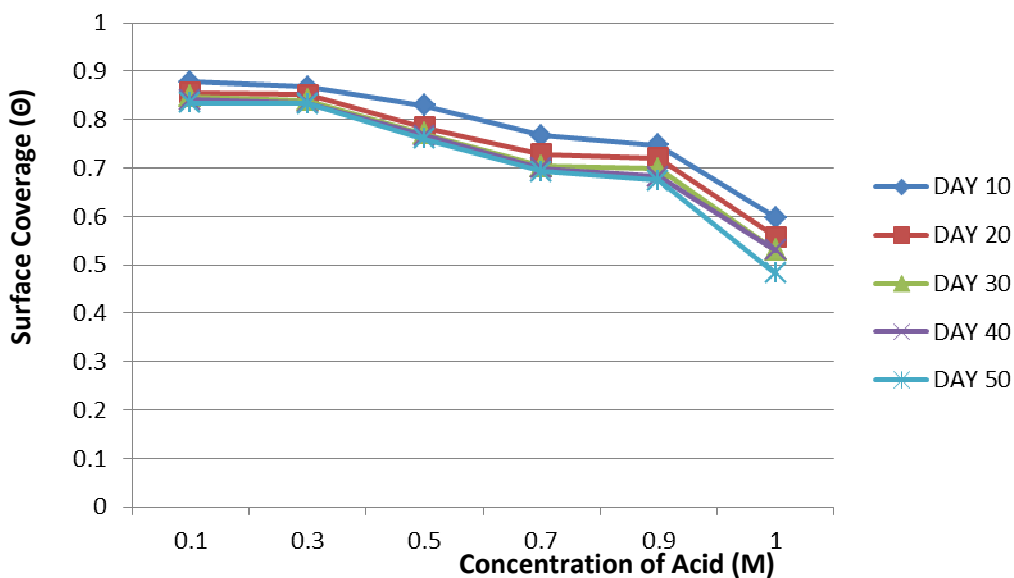


Figure 11: Graph of Degree of Surface Coverage (Θ) Vs Acid Conc. (M)

The **Figure 11** shows the variation of the Degree of Surface Coverage (Θ) with various concentration of the H_2SO_4 recorded for alloy 304L in 0.001M Adeninesolution at 10 days interval for 50days

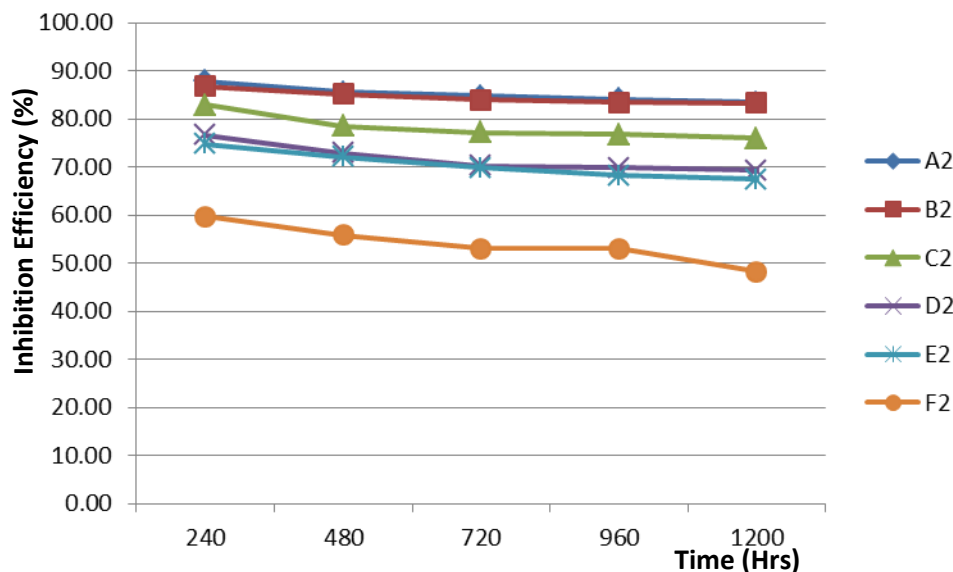


Figure 12: Graph of Inhibition Efficiency (%) Vs Time (hrs)

The **Figure 12** shows the variation of the Inhibition Efficiency (%) with the immersion time (hrs) recorded for alloy 304L in 0.001M Adeninesolution as inhibitor with various H_2SO_4 concentrations. (A₂) 0.1M H_2SO_4 ; (B₂) 0.3M H_2SO_4 ; (C₂) 0.5M H_2SO_4 ; (D₂) 0.7M H_2SO_4 ; (E₂) 0.9M H_2SO_4 ; (F₂) 1.0M H_2SO_4

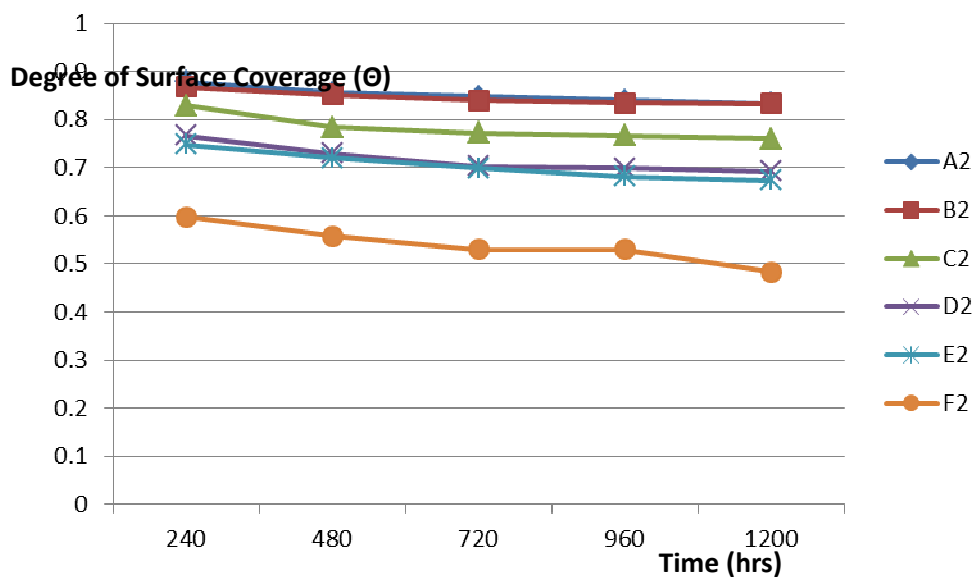


Figure 13: Graph of Degree of Surface Coverage (Θ) Vs Time (hrs)

The **Figure 13** shows the variation of the Degree of Surface Coverage (Θ) with the immersion time (hrs) recorded for alloy 304L in 0.001M Adeninesolution as inhibitor at various H_2SO_4 concentrations. (A₂) 0.1M H_2SO_4 ; (B₂) 0.3M H_2SO_4 ; (C₂) 0.5M H_2SO_4 ; (D₂) 0.7M H_2SO_4 ; (E₂) 0.9M H_2SO_4 ; (F₂) 1.0M H_2SO_4

DISCUSSION

The graph of **Figures 2 and 3** show the weight loss of UNS S304L in varied concentration of sulphuric acid solutions without adenine added and with adenine (as inhibitor) added.

Also, the graph of **Figures 4 and 5** show the weight loss with time of AISI 304L of the uninhibited and the inhibited samples. It can be seen that the weight loss increased with increase in the concentration of the sulphuric acid in both the uninhibited and the inhibited samples. But when compared, the amount of weight loss recorded by the

uninhibited samples far exceeded the weight loss recorded by the inhibited samples. It is also observed that the weight loss increased with time in both the uninhibited and the inhibited samples.

More so, the slope of each line (weight loss per unit time; $\text{mg}/\text{cm}^2\text{h}$) of **Figures 4 and 5** represent the corrosion rate of alloy 304L at the specified conditions. In addition, the graph of **Figures 6 and 7** shows the corrosion rate of AISI 304L in varied concentration of the Tetraoxosulphate VI acid solutions without adenine and with adenine as inhibitor present. From the **Figures 3 and 7**, it can be seen that the adenine is an effective inhibitor for the corrosion of AISI 304L in sulphuric acid solutions as the corrosion of the AISI 304L was retarded by the presence of 0.001M concentration of adenine solution [17, 25]. Thus, the continuous decrease in corrosion penetration rate suggests that a protective film with inhibitive property is formed on the metal/environment interface. This implies that the inhibition mechanism involves the adsorption of protonated adenine molecules onto the 304L surface

The **Figures 2 and 6** also show the effect of the sulphuric acid concentration on the corrosion of the 304L. It can be observed that the corrosion penetration rate of AISI 304L increased with increase in the concentration of the sulphuric acid within the concentration range of the acid investigated in this work. This is in agreement with the works of Iliyasa *et al* and Abdallah [5, 6]. However, the uninhibited samples experienced a higher corrosion rate compared with the inhibited samples which showed a lower corrosion rate due to the presence of the 0.001M concentration of the Adenine.

Nevertheless, from **Figure 8 and 9** it can be noticed that the corrosion penetration rate of both the uninhibited and inhibited samples of alloy 304L decreased with increase in immersion time. According to Loto and Adesomo [23], the decrease in corrosion penetration rate with time can be attributed to an initial chemical reactivity of anodic dissolution at the metal/solution interface within the first 24 hours of the experiment, and that the continued decline of corrosion penetration rate with time could be due to the aggressiveness of the chemical reactivity, transport properties of the environment, concentration of corrosion species in the medium, pH and the temperature of the corrosion medium which decrease as time is appreciably increased.

Similarly, the **Figures 10 and 11** show the influence of increasing the concentration of the sulphuric acid on the inhibition efficiency and degree of surface coverage during the corrosion inhibition of AISI 304L in varied concentration of sulphuric acid solutions with 0.001M Adenine concentration as inhibitor. From the figures (**Figures 10 and 11**) it can be seen that both the inhibition efficiency and the degree of surface coverage of the inhibitor (0.001M Adenine) decreased as the concentration of the sulphuric acid was increased showing a reduction in inhibition efficacy of the said inhibitor. The decrease in inhibition efficiency and degree of surface coverage of the 0.001M Adenine concentration was remarkably significant at higher concentration of the sulphuric acid with an inhibition efficiency and degree of surface coverage of 87.86% and 0.8786 at 0.1M sulphuric acid solution, 86.75% and 0.8675 at 0.3M sulphuric acid solution, 82.89% and 0.8289 at 0.5 M sulphuric acid solution, 76.63% and 0.7663 at 0.7M sulphuric acid solution, 74.75% and 0.7475 at 0.9M sulphuric acid solution, and , 59.72% and 0.5972 at 1.0M sulphuric acid solution after 240 hours (10 days). Thus, the increasing concentration of the sulphuric acid solution had marked influence on the inhibiting effect (action) of the 0.001M Adenine during the corrosion inhibition of AISI 304L in varied concentration of sulphuric acid.

Moreover, from **Figures 12 and 13**, it is revealed that both the inhibition efficiency and degree of surface coverage reduced with increase in immersion time. According to Ijeoma [24] and Sastri [19] this observation could be due to the inhibition reacting with contaminants and other components of the system (such as corrosion products) leading to the loss of inhibitor or the inhibitor getting depleted/used-up with time due to chemical reactions occurring within the system. The inhibitor can also be lost due to film formation.

CONCLUSION

The results showed that adenine is a safe inhibitor; it is environmentally friendly and poses no serious threat to life. The adenine has good inhibition effect for the corrosion of AISI 304L austenitic stainless steel in varied sulphuric acid solutions, and, inhibition efficiency and degree of surface coverage of the adenine both decreased with increase in sulphuric acid concentration as well as with the immersion time. Thus, the increasing concentration of the sulphuric acid solution had marked influence on the inhibiting effect (action) of the 0.001M Adenine concentration during the corrosion of AISI 304L in varied concentration of sulphuric acid. The corrosion rate of AISI 304L increased with increase in sulphuric acid concentration but decreased with immersion time.

REFERENCES

- [1] Onyekpe, B.O. Corrosion in Oil and Gas production. Ambik Press, Benin City, 2002.

- [2] DeGiorgi, V.G, Corrosion Basics and Computer Modelling, Naval Research Laboratory, D.C.203755000, U.S.A.,**1993**,pp 48.
- [3]Seifedine, K. (2008). European Journal of Scientific Research, ISSN 1450-216X Vol. 22 No.4, **2008**, 508-516.
- [4] Khatak, H.S and BaldevRaj. Corrosion of Austenitic Stainless Steels: Mechanism, Mitigation and Monitoring, Woodhead Publishing Li, **2002**.
- [5] Iliyasu, I, Yawas, D.S and Aku, S.Y. *Journal of Advances in Applied Sciences Research*, **2012**, 3(6): 3909-3915
- [6] Abdallah, M. *Journal of Materials Chemistry and Physics*, **2003**, Vol. 82, Pg 782-786
- [7] Gad Allah, A.M., Hefny. M.M., Salih, S.A., El-Basiouny, M.S. *Corrosion Science*, **1989**, Vol. 45, No. 7.
- [8] Amitha Rani, B.E., and Bai J. Basu. Green Inhibitors for Corrosion Protection of Metals and Alloys: An Overview, *International Journal of Corrosion*, **2012**.
- [9] Bouklah M., Hammouti B., Aouniti A and Benhadda T. *Journal of Applied Electrochemistry*, **2005**, Vol. 35, No. 11, pp1101.
- [10] Chikkur B. Pradeep Kumar and Kikkeri N. Mohana, The Effect of Achyranthesaspera Extracts on Mild Steel Corrosion in Industrial Water Medium, *ISRN Corrosion*, Vol. **2013**.
- [11] El-Hajjaji F, Belkhmima R.A, Zerga, Sfaira M, Taleb M, Ebn M. Touhami, and Hammouti B, *J. Mater. Environ. Sci.* **2014**, Vol. 5, Pg 263-270
- [12] Aksiit A.A., and Biligic S. *Corrosion Science*, **1992**, Vol. 33, Pg 379
- [13] Moretti, G., and Guidi, F. Tryptophan as Copper corrosion Inhibitor in 0.5M aerated sulphuric acid, Department of Chemistry, University of Venice, Italy, **2002**
- [14] Scendo, M. *Corrosion Science*, **2008**, Vol. 50, Pg 2070-2077
- [15] Scendo, M. *Corrosion Science*, **2007**, Vol. 49, Pg 2985
- [16] Scendo, M. *Corrosion Science*, **2007** Vol. 49, Pg 3953
- [17] Scendo, M. *International Journal of Electrochemical Science*, **2013** Vol. 8, Pg 9201-9221
- [18] Sayed S. Abdel Rehim, Omar A. Hazzaxi, Mohammed A. Amin, Khaled F. Khaled. *Corrosion Science*, **2008**, Vol. 50, Pg 2258-2271
- [19] Sastri, V.S. Green Corrosion Inhibitors: Theory and Practice, 1st Ed. John Wiley and Sons, Inc. New York, **2011**.
- [20] Chitra S., Parameswari K., Vidhya M., Kalishwari M and Selvaraj A. *International Journal of Electrochemical Science*, **2011**, Vol. 6, Pg 4593-4613.
- [21] HabibatMomoh-Yahaya, NnabukO.Eddy, Johnson F. Iyun, Casmir E. Gimba and Emeka E. Oguzie. *Journal of materials Science Research*, **2013**, Vol.2, No. 1.
- [22] Fontana MG, Greene ND, "Corrosion Engineering." McGraw-Hill International Book Co., 2nd Edition, **1978**, 194, 261.
- [23] Loto, C.A and Adesomo, M.A. *African Journal of Science and Technology*, **1988**, Series A Vol.7, No. 1
- [24] Ijeoma, M.N.C. Elements of Corrosion and Protection Theory. Auto Century Publication Co. Ltd, Enugu, Nigeria, **1991**.
- [25] Ofuyekpone, O, Effect of Adenine as Corrosion Inhibitor on the Corrosion of Alloy 304L in 1.0 M Sulphuric Acid Solution. M.Eng. Dissertation, Department of Mechanical Engineering, University of Benin (Benin City, Nigeria, **2014**).