

Full Length Research Paper

Inhibitive action of hexadecyltrimethylammonium bromide (HDTAB) on aluminium alloy (AA3003) corrosion in hydrochloric acid medium

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The inhibitive effect of hexadecyltrimethylammonium bromide (HDTAB) on the corrosion of aluminium alloy (AA3003) in 1 M hydrochloric acid medium was studied by weight loss method in relation to the temperature (25 to 55°C) and inhibitor concentration range 5.10^{-4} to 10^{-2} M. This study showed that the inhibitor adsorbs on aluminium alloy (AA 3003) according to the modified Langmuir adsorption isotherm; the inhibitor efficiency decreases with increasing temperature. Adsorption free energies (ΔG°_{ads}) and apparent activation energies (E_a) were determined. The Adsorption free energies reveal that the adsorption process is spontaneous. Furthermore, some adsorption thermodynamic parameters (ΔH°_{ads} , ΔS°_{ads} , Q_{ads}) were estimated. Phenomenon of physical adsorption was proposed from the thermodynamic functions of adsorption and activation parameters obtained.

Key words: Corrosion, inhibitor, adsorption, desorption, free adsorption energy, activation energy.

INTRODUCTION

Corrosion of metals and alloys is a common phenomenon in industry and it has received a considerable amount of attention. Aluminium and its alloys are important materials with wide range of industrial applications. The inhibition of their corrosion is a subject of technological importance due to the increase industrial applications of these materials (Li et al., 2008; Saliyan et al., 2008; Tang et al., 2006). Aluminium is considered as an advanced material in many industrial processes due to its relatively low cost, high strength and high corrosion resistance (Wen et al., 1987). This metal is exposed to the action of acids in industrial processes where acids play important roles such as in oil well acidizing, acid pickling, acid cleaning and descaling (Keles et al., 2008). These processes are generally accompanied by considerable dissolution of the metal, so arises the problem of the material protection.

The use of corrosion inhibitors is a current practice in industry (Abdallah, 2003; Armstrong et al., 1994). These corrosion inhibitors are chemical compounds usually used in small concentrations whenever a metal is in

contact with an aggressive medium. The presence of such compounds retards the corrosion process and keeps its rate to a minimum and thus prevents economic losses due to metallic corrosion. The chemical compounds that could be used for this purpose may be organic or inorganic.

However, not just any chemical compound can be used as a corrosion inhibitor. There are some requirements that the compound must fulfil. Regarding the chemical structure and chemical behaviour, an inorganic compound must be able to oxidize the metal, forming a passive layer on its surface. On the other hand, a molecule of an organic compound must have features that give it the ability to act as corrosion inhibitor. Apart from the structural aspect, there are also environmental considerations. During several years compounds on base of chromium (IV) were used as inhibitors of the corrosion of aluminium and its alloys (Bazzi et al., 2002). Regrettably, these compounds were found to be carcinogenic and toxic (Stoyanova et al., 1997). Consequently, an intense research effort is being undertaken to replace chromate compounds by more ecological compounds in anti-corrosion aluminium treatment (Manov et al., 2000) which explains the current use of organic inhibitors containing unsaturated bounds and / or polar atoms (Aksüt et al., 1997; Rengazani, 1994;

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Stupnisek-Lisac et al., 1995; Szeptycka, 1995).

The inhibitive power of the organic inhibitors has been interpreted in term of many different characteristics such as molecular size, molecular weight, molecular structure and the nature of heteroatom present in the molecule (Muralidharan et al., 1995; Quraishi et al., 2001). Nitrogen containing compounds have been found to serve as good inhibitors of corrosion (Popova et al., 2007; Granese et al., 1992; Subramanian et al., 1990). Generally, it has been assumed that the first stage in the action mechanism of the inhibitors in aggressive acidic media is the adsorption of the inhibitors onto the metal surface. The processes of adsorption of the inhibitors depend mainly on certain physico-chemical properties of the inhibitor molecule such as electron density at the donor atom, π -orbital character and electronic structure of the molecule (Emregul et al., 2004).

Several authors (Ovari et al., 1988; Tomcsanyi et al., 1989; Frers et al., 1990) have studied the corrosion of aluminium and its alloys and their inhibition by organic compounds in acid solutions. Various aliphatic and aromatic amines as well as nitrogen heterocyclic compounds were studied as corrosion inhibitors for pure aluminium in acid media (Hackerman et al., 1958; Hackerman et al., 1984). Hydrazine compounds (Isaa et al., 1973), organic acid and their salt (Müller, 2004) were also found to inhibit the corrosion of aluminium in HCl solutions.

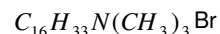
The aim in the present investigation is to examine the inhibitive action of the hexadecyltrimethylammonium bromide (HDTAB) towards the corrosion of aluminium alloy (AA3003) in 1 M hydrochloric acid solution. In this work, we have estimated the corrosion parameters by weigh loss of the metal material after 1 h of immersion in the acidic medium. No previous studies have reported the use of this molecule to inhibit the corrosion of aluminium alloy in the studied medium.

MATERIALS AND METHODS

Our experimental device includes essentially a thermostatic bath (SELECTA) which was used to maintain the temperature, a thermometer to control the temperature, a chronometer to estimate the time and an analytical electronic METLER balance (precision $\pm 0.1\text{mg}$).

Materials

Our sample was a rod of aluminium alloy (AA 3003) measuring 1.2 cm in length and 0.3 cm in diameter. The chemical composition of the aluminium alloy is Mn-1.32% Fe- 0.45% Cu-0.11 % Si- 0.23% Mg- 0.02 % Cr- 0.01% Zn- 0.01 % and the remainder being Al. The chemical structure of hexadecyltrimethylammonium bromide (HDTAB) from MERCK is given below:



The compound was a white powder with molecular weight, $364.5 \text{ g. mol}^{-1}$.

Weight loss experiments

The samples were polished successively with fine grade emery papers, cleaned with acetone, washed with double distilled water and dried in moisture free desiccators. Tests were conducted under total immersion conditions in 50 mL of test solutions maintained at (25 to 55 °C). The pre-cleaned and weighed samples were in the beaker containing the test solution. All tests were made in aerated solutions and were run triplicate to guarantee the reliability of the results. To determine the weight loss at the end of the test, the samples were retrieved from tests solutions after 1 h, washed in 65% nitric acid for 5 s to remove the corrosion products, using bristle brush and rinsed with double distilled water, dried and reweighed. The weight loss was taken as the difference between the initial weight and the weight after 1 h immersion time.

Analytical reagent grade HCl from MERCK ($P = 37\%$, $d = 1.19$, $M = 36.49 \text{ g. mol}^{-1}$) and double distilled water were used to prepare tests solutions of 1 M HCl for all experiments.

The corrosion rate W in ($\text{g. cm}^{-2} \cdot \text{s}^{-1}$) was calculated from the following relation:

$$W = \frac{m_1 - m_2}{St} \quad (1)$$

Where m_1 is the weight of the sample before immersion in gram, m_2 is the weight of the sample after immersion in gram, S is the total area in contact with the liquid in cm^2 and t is the immersion time in s. The surface coverage θ was calculated from the corrosion rate as follows:

$$\theta = \frac{W_0 - W}{W_0} \quad (2)$$

Where W_0 is the corrosion rate without inhibitor and W is the corrosion rate in presence of the inhibitor. The inhibition efficiency (% IE) of the organic molecule is then:

$$\% \text{ IE} = \frac{W_0 - W}{W_0} * 100 \quad (3)$$

RESULTS AND DISCUSSION

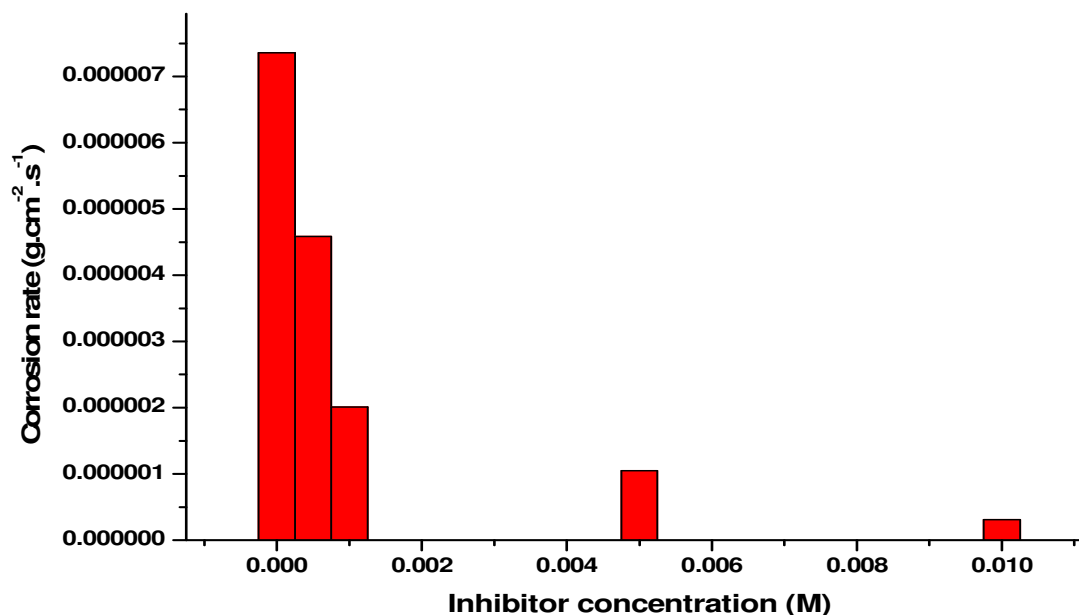
Effect of the inhibitor concentration

Table1 gives the corrosion parameters of aluminium alloy (AA 3003) at $T = 35^\circ \text{C}$ for a solution of 1 M HCl (blank) and solutions of 1 M HCl containing hexadecyltrimethylammonium bromide (HDTAB). The calculated values of corrosion rate, using Equation (1) as well as inhibition efficiency values, evaluated using Equation (2) are collected in this table. Figure 1 shows a representative plot of the variation of the corrosion rate of the sample with the inhibitor concentration.

It can be seen that corrosion rate is reduced in the presence of hexadecyltrimethylammonium compared to in its absence (blank). The addition of HDTAB to

Table 1. Corrosion parameters for blank and solutions containing different concentrations of HDTAB at T = 35 °C.

| C (M) | m ₁ (g) | m ₂ (g) | Δm (g) | W (g.cm ⁻² .s ⁻¹) | IE (%) | θ |
|--------------------|--------------------|--------------------|--------|--|--------|-------|
| 0 | 0.2607 | 0.2270 | 0.0337 | 7.3576.10 ⁻⁶ | – | – |
| 5.10 ⁻⁴ | 0.2606 | 0.2396 | 0.0210 | 4.5849.10 ⁻⁶ | 37.68 | 0.377 |
| 10 ⁻³ | 0.2607 | 0.2515 | 0.0092 | 2.0090.10 ⁻⁶ | 72.70 | 0.727 |
| 5.10 ⁻³ | 0.2608 | 0.2560 | 0.0048 | 1.0479.10 ⁻⁶ | 85.76 | 0.857 |
| 10 ⁻² | 0.2607 | 0.2593 | 0.0014 | 3.0566.10 ⁻⁷ | 95.85 | 0.958 |

**Figure 1.** Variation of corrosion rate of aluminium alloy (AA3003) with inhibitor concentration at T = 35 °C.

hydrochloric acid solutions retards the corrosion of aluminium alloy (AA 3003) and the extent of retardation is concentration dependent. Similar observations have been seen in the literature (Obot et al., 2009). Inhibition efficiency as given in Table 1 is found to increase with increasing concentration (Figure 2). This can be explained by the adsorption of the studied molecule forming a layer film (Riggs, 1973).

Effect of temperature

Table 2 gives the corrosion rate, the surface coverage and the inhibition efficiency for different temperatures. The results listed in this table show that the corrosion rates of aluminium alloy (AA 3003) is temperature dependant and it increases with increasing temperature. Figure 3 allows comparing the dissolution rate of aluminium alloy (AA 3003) in the blank and that in hydrochloric acid solutions containing the inhibitor. Figure 3 shows clearly that the use of hexadecyltrimethylammonium bromide (HDTAB) reduces

considerably the corrosion rate of aluminium alloy (AA 3003) in spite of the rise of temperature.

Figure 4 shows a representative plot of the variation of inhibition efficiency with the temperature. It can be observed that corrosion efficiency is temperature dependant. The higher the temperature, the lower is the inhibition efficiency. Thus we can see that the efficiency which is 97.2 at 25°C is lowered down to 63.4 at 55°C. Decrease in inhibitor efficiency with increasing temperature (Obot et al., 2009) may be attributed to increase in the solubility of the protective film and of any reaction products precipitated on the surface of Aluminium alloy (AA 3003) that may otherwise inhibit the corrosion process.

The decrease in inhibition efficiency with increasing temperature as observed in Figure 4 is suggestive of physisorption mechanism generally attributed to electrostatic interaction between charged molecules and charged metal. This observation can be interpreted by the reduction in stability of the adsorbed film at higher temperature. According to Umoren et al., 2009, as temperature increases, Gibbs free energy and change in

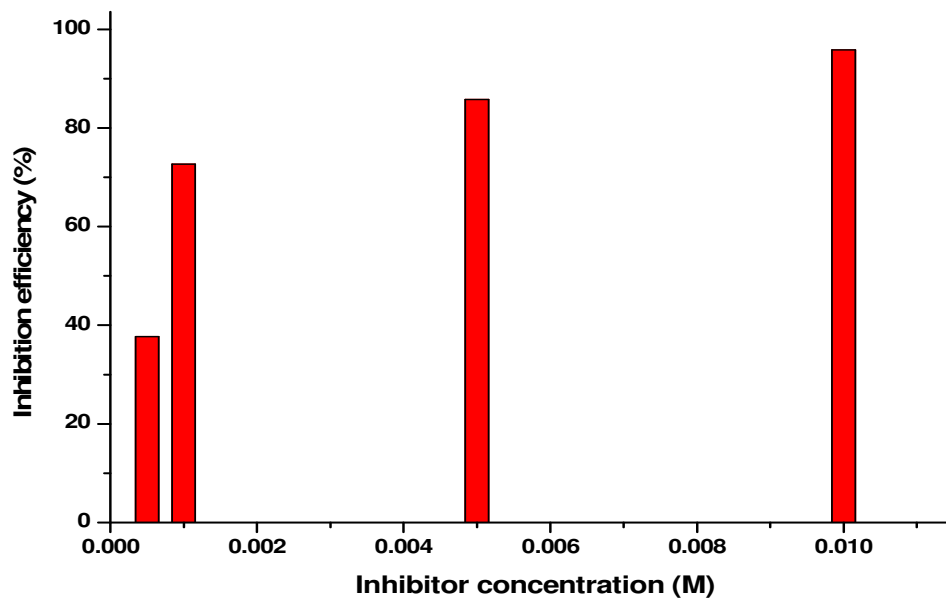


Figure 2. Variation of Inhibition efficiency of HDTAB with inhibitor concentration at T = 35 °C.

Table 2. Evolution of corrosion rate of aluminium alloy (AA 3003) with temperature in 1 M hydrochloric solution in the absence and presence of the inhibitor (C = 10⁻² M).

| T (°C) | Δm_{Blank} (g) | Δm_{inh} (g) | W_0 (g.cm ⁻² .s ⁻¹) | W (g.cm ⁻² .s ⁻¹) | θ | IE (%) |
|--------|-------------------------------|-----------------------------|--|--|----------|--------|
| 25 | 0.0036 | 0.0001 | $7.859 \cdot 10^{-7}$ | $2.184 \cdot 10^{-8}$ | 0.972 | 97.2 |
| 35 | 0.0274 | 0.0022 | $4.901 \cdot 10^{-6}$ | $4.803 \cdot 10^{-7}$ | 0.902 | 90.2 |
| 45 | 0.0406 | 0.0086 | $8.864 \cdot 10^{-6}$ | $1.878 \cdot 10^{-6}$ | 0.788 | 78.8 |
| 55 | 0.0560 | 0.0205 | $1.223 \cdot 10^{-5}$ | $4.476 \cdot 10^{-6}$ | 0.634 | 63.4 |

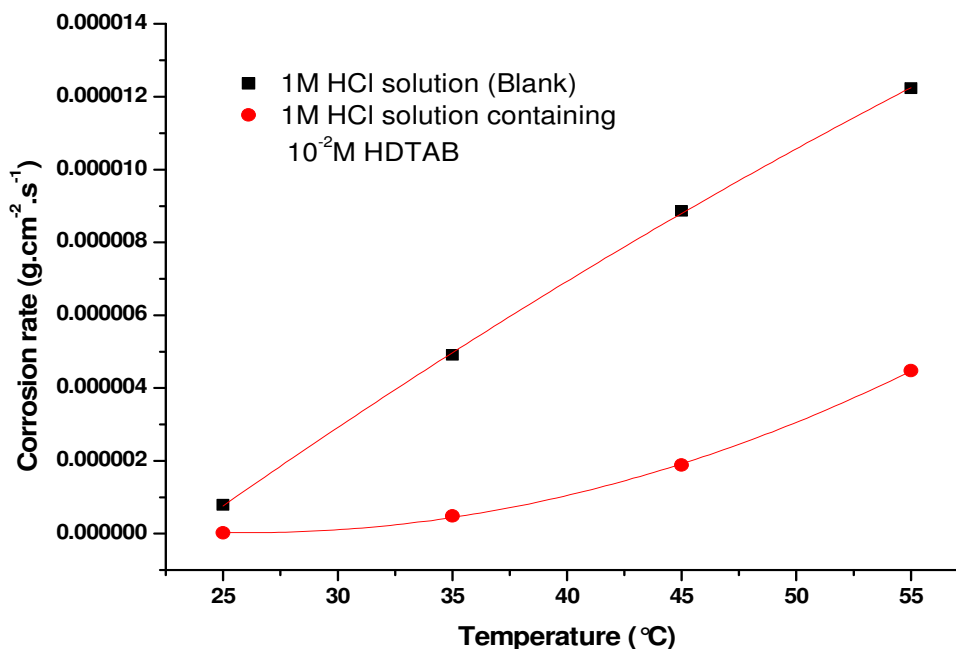


Figure 3. Variation of the corrosion rate with the temperature in 1.0 M HCl and 1 M HCl containing 10⁻² M HDTAB.

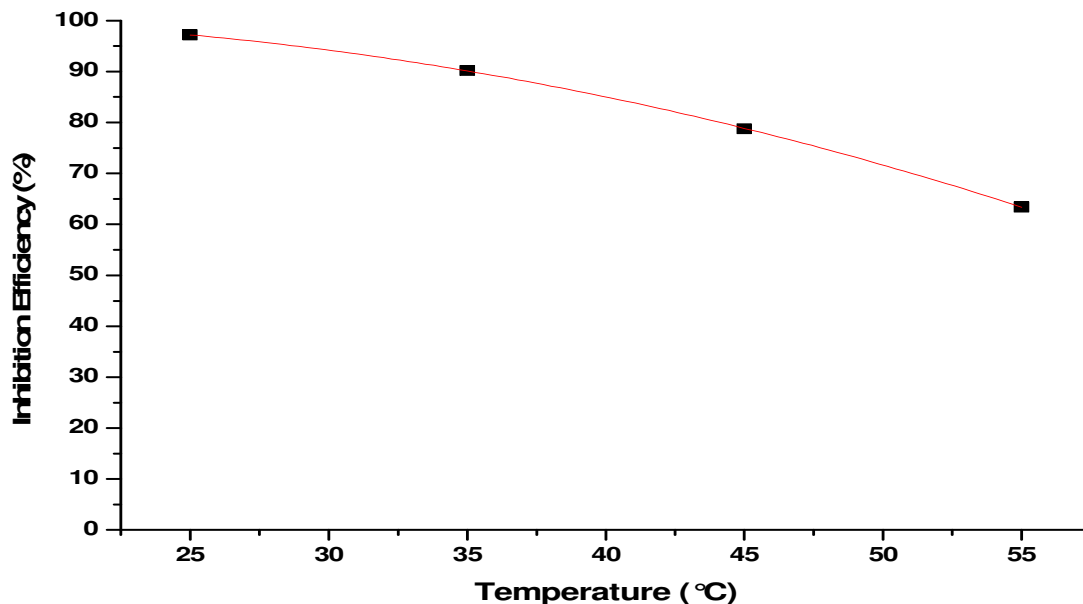


Figure 4. Variation of the Inhibition efficiency with the temperature in 1.0 M HCl containing 10^{-2} M HDTAB.

enthalpy rise to a higher value, so that some of the chemical bonds joining the molecules onto the metallic surface are impaired and the film stability reduced. It may further be attributed to a possible shift of the adsorption/desorption equilibrium towards desorption of adsorbed inhibitor due to increasing solution agitation. Thus, as the temperature increases the number of adsorbed molecules decreases, leading to a decrease in the inhibition efficiency.

Adsorption isotherm

Adsorption isotherms provide information about the interaction of the adsorbed molecules with the metal surface (Naderi et al., 2009). The adsorption of an organic adsorbate at metal-solution interface can be presented as a substitution adsorption process between the organic molecules in aqueous solution and the water molecule on metallic surface:



Where $\text{Org}_{(\text{sol})}$ and $\text{Org}_{(\text{ads})}$ are the organic species dissolved in the aqueous solution and adsorbed onto the metallic surface, respectively; $\text{H}_2\text{O}_{(\text{ads})}$ and $\text{H}_2\text{O}_{(\text{sol})}$ are the water molecule adsorbed onto the metallic surface and in the bulk solution; x is the size ratio representing the number of water molecule replaced by one organic adsorbate.

Adsorption depends mainly on the charge and nature of the metal surface, electronic characteristics of the metal surface, adsorption of solvent, other ionic species and

temperature of corrosion reaction. Adsorption of the inhibitor involves the formation of two types of interactions responsible for bonding inhibitor to the metal surface. The first type, physical adsorption is weak indirect interaction and is due to electrostatic attraction between inhibiting organic ions or dipoles and the electrically charged surface of the metal. The second type of interaction, chemical adsorption occurs when direct forces govern the interaction between adsorbate and adsorbent. Chemical adsorption involves charges charing or charge transfer from adsorbate to the metal surface atoms in order to form a coordinate type bond. Chemical adsorption has a free energy of adsorption higher than physical adsorption and hence, usually it is irreversible (Trabanelli, 1987).

The establishment of a corrosion inhibitor adsorption isotherm can provide important clues to the nature of the metal-inhibitor interactions. In order to obtain the adsorption isotherm, linear relation between the surface coverage θ and the inhibitor concentration must be found. Attempts were made to fit the θ values to various isotherms including Langmuir, Temkin, Frumkin and Freundlich. The best fit was obtained with the Langmuir isotherm. This adsorption isotherm is described by the following equation:

$$\frac{C}{\theta} = \frac{1}{K_{\text{ads}}} + C \quad (5)$$

Where θ is the degree of surface coverage, C is the molar inhibitor concentration in the bulk solution and K_{ads} is the equilibrium constant of the adsorption process. The

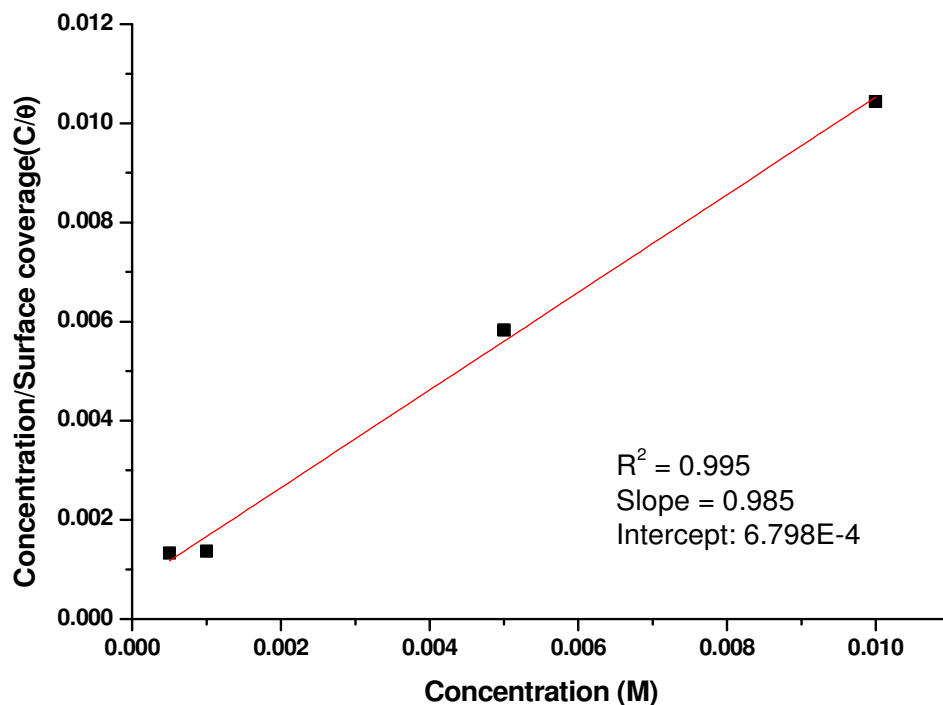


Figure 5. Curve fitting of the weight loss data of aluminium alloy (AA 3003) in 1.0 M HCl solution containing 10^{-2} M HDTAB at $T = 35^\circ\text{C}$.

corresponding isotherm is obtained by plotting C/θ versus C (Figure 5).

The linear relationship of C/θ with C shows that the adsorption of HDTAB obeys the Langmuir adsorption isotherm. In our case, the line has a slope of 0.985 and the correlation coefficient (R^2) is 0.995. This is indicative of some divergence from pure monolayer adsorption and can be attributed to interactions between adsorbate species on the metal surface as well as changes in the adsorption heat with increasing surface coverage (Obot et al., 2009), factors which were not taken into consideration in derivation of this isotherm. The deviation of the slope from unity is often interpreted as a sign that the adsorbing species occupy more or less a typical adsorption site at the metal/solution interface (Hosseini et al., 2003). The Langmuir adsorption isotherm cannot be applied rigorously. A modified Langmuir adsorption isotherm (Ali et al., 2003), can be applied to this phenomenon which is given by the corrected equation:

$$\frac{C}{\theta} = \frac{n}{K_{ads}} + nC \quad (6)$$

Adsorption free energy

The equilibrium constant for the adsorption process from Langmuir isotherm is related to the standard free energy of adsorption by the expression:

$$K_{ads} = \frac{1}{55.5} \exp\left(-\frac{\Delta G_{ads}^\circ}{RT}\right) \quad (7)$$

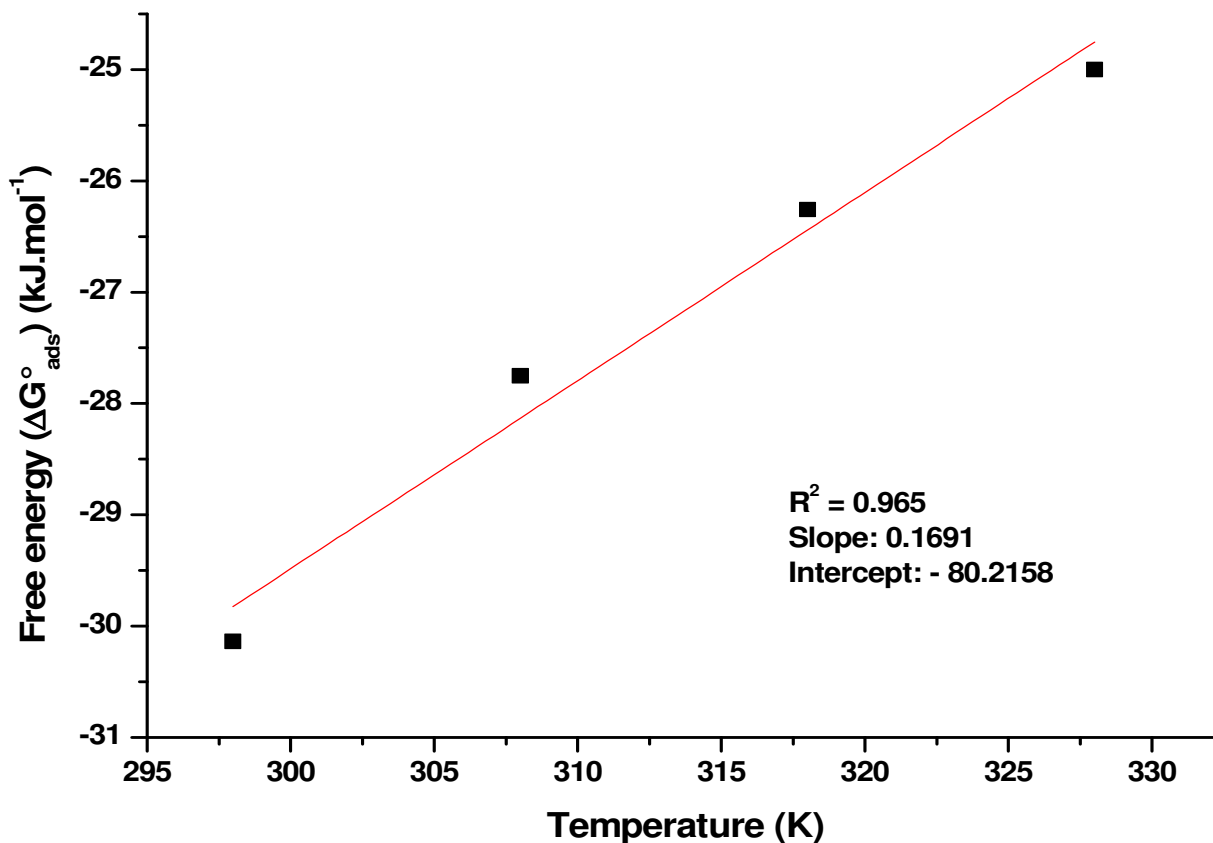
Where R is the molar gas constant, T , the absolute temperature and the value 55.5 is the concentration of water in solution expressed in $\text{mol}\cdot\text{L}^{-1}$. The standard adsorption free energy change ΔG_{ads}° which can characterize the interaction of adsorbed molecules and metal surface was calculated, using Equations (6) and (7). The obtained values are listed in the Table 3. The negative values of ΔG_{ads}° ensure the spontaneity of the adsorption process and the stability of the adsorbed layer on the aluminium alloy (AA 3003) surface. Generally, the values of ΔG_{ads}° around $-20 \text{ kJ}\cdot\text{mol}^{-1}$ or less negative are consistent with physisorption, while those around $-40 \text{ kJ}\cdot\text{mol}^{-1}$ or more negative involve chemisorption (Quraishi et al., 2005). The values found for HDTAB on the aluminium alloy indicate that the adsorption is physisorption (electrostatic interactions) and probably no covalent bonds exist between the inhibitor molecules and the metal surface. The relation between ΔG_{ads}° , ΔH_{ads}° and ΔS_{ads}° is given by the following basic equation:

$$\Delta G_{ads}^\circ = \Delta H_{ads}^\circ - T\Delta S_{ads}^\circ \quad (8)$$

The change in enthalpy ΔH_{ads}° and entropy ΔS_{ads}° of the

Table 3. Calculated thermodynamic adsorption parameters from Langmuir adsorption isotherm at inhibitor concentration $C = 10^{-2}$ M.

| T (K) | K_{ads} (L.mol ⁻¹) | ΔG_{ads}° (kJ.mol ⁻¹) | ΔH_{ads}° (kJ.mol ⁻¹) | ΔS_{ads}° (J.mol ⁻¹ .K ⁻¹) |
|-------|----------------------------------|--|--|--|
| 298 | 3471.4 | -30.14 | -80.2 | -169.1 |
| 308 | 920.4 | -27.75 | - | - |
| 318 | 371.7 | -26.26 | - | - |
| 328 | 173.2 | -25.00 | - | - |

**Figure 6.** Variation of free adsorption energy with temperature in 1.0 M HCl containing 10^{-2} M HDTAB.

adsorption process can be obtained by plotting ΔG_{ads}° versus temperature (Figure 6).

The straight line was obtained with a slope equals to $(-\Delta S_{ads}^{\circ})$ and an intercept equals to (ΔH_{ads}°) . The values of the change in adsorption enthalpy and entropy are listed in Table 3. The negative sign of ΔH_{ads}° indicates that the adsorption of the inhibitor molecules is an exothermic process. The value of ΔS_{ads}° in the presence of HDTAB is negative, meaning that a decrease in disordering takes place on going from reactants to the metal adsorbed species reaction complex (Kissi et al., 2006).

Activation energy

The relationship between the temperature, the inhibition efficiency (%IE) of an inhibitor and the activation energy (E_a) in the presence of an inhibitor was given as follow (Popova et al., 2003):

1. Inhibitors whose (%IE) decreases with increasing temperature; the value of the activation energy (E_a) found is greater than that in the uninhibited solution.
2. Inhibitors whose (%IE) does not change with temperature variation; the activation energy (E_a) does not change in the presence or absence of inhibitors.
3. Inhibitors whose (%IE) increases with increasing

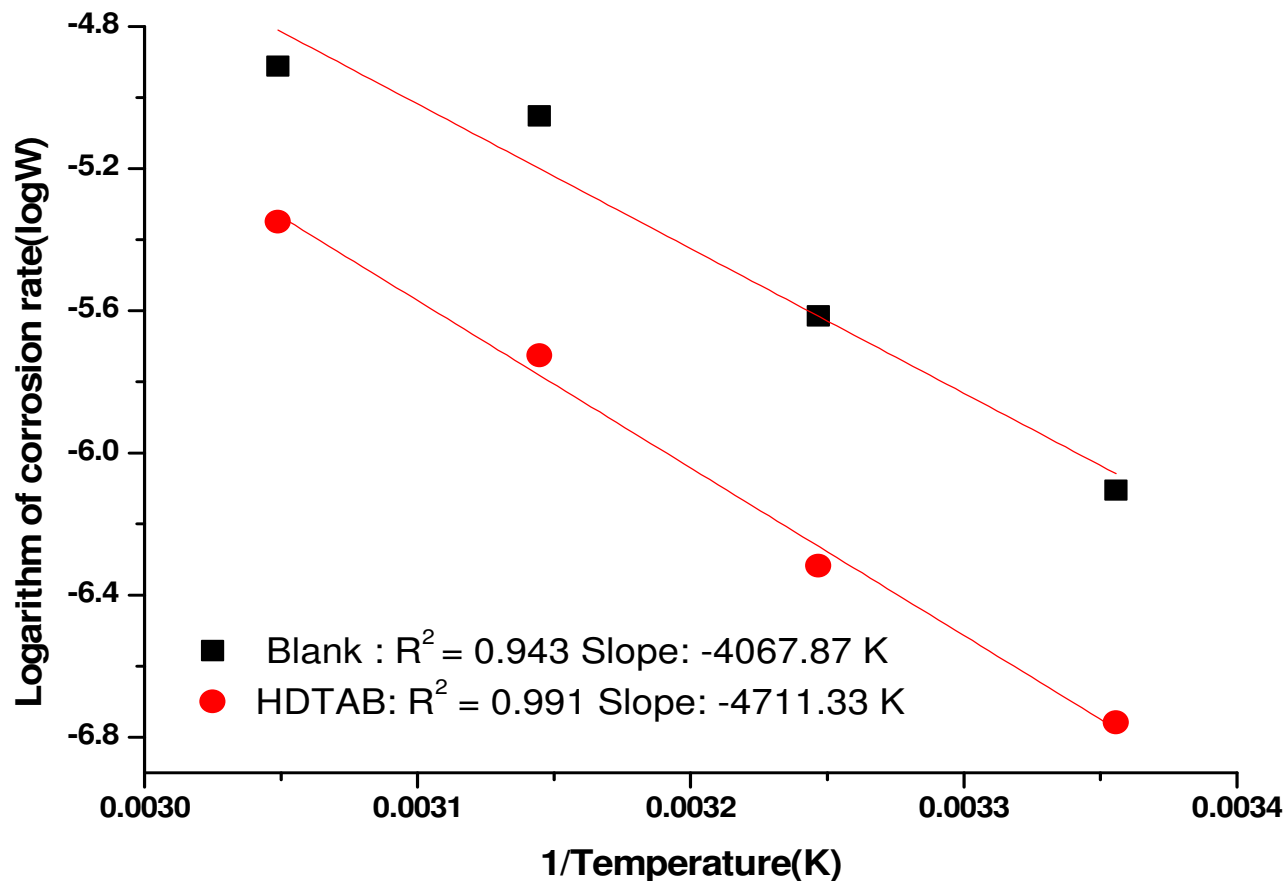


Figure 7. Arrhenius plots of corrosion Log W versus 1/T of 1.0 M HCl and 1.0 M HCl containing 10^{-2} M HDTAB.

temperature; the value of activation energy (E_a) found is less than that in the uninhibited solution.

In order to specify the type of adsorption, the value of the activation energy (E_a) was calculated with the help of the Arrhenius equation (Tang et al., 2005):

$$W = k \exp\left(-\frac{E_a}{RT}\right) \quad (9)$$

Where W is the corrosion rate, k the preexponential factor, E_a the activation energy, R the constant of perfect gases and T, the absolute temperature. Plotting log W versus 1/T (Figure 7) for the blank and for the solution containing HDTAB, leads to the values of E_a .

The activation energies deduced from the slopes of the straight lines functions shown in Figure 7 are $E_a = 77.8$ kJ. mol $^{-1}$ (Blank) and $E_a' = 90.1$ kJ. mol $^{-1}$ in presence of the inhibitor. The higher value of the activation energy (E_a) of the process in the presence of an inhibitor when compared to that in its absence (Umoren, 2008) is attributed to its physical adsorption; its chemisorption is pronounced in the opposite case. In our investigations, the activation energy found in presence of HDTAB is

higher than that obtained in its absence, so the adsorption of the studied molecule onto the aluminium alloy (AA 3003) surface is found to be physical in nature.

Adsorption mechanism

The value of pH_{zch} , which is defined as the pH at point of zero charge, is equal to 9.1 for aluminium (Yurt et al., 2006). So the aluminium alloy (AA 3003) surface is positively charged for the pH of 1.0 M HCl solutions used in these investigations. The anions could be specifically adsorbed and create an excess negative charge towards the solution and favoured adsorption of the cation. Indeed it is known that halide ions present in an inhibiting solution adsorb on corroding metal surface by creating oriented dipoles and facilitate the adsorption of the inhibitor cations on the dipole (Bentiss et al., 2000; Oguzie et al., 2007). In our case, adsorption occurs via electrostatic interaction between a negatively charged aluminium surface, which is provided with a specifically adsorbed anion (Cl^- , Br^-) on aluminium with the positively charged species [$\text{C}_{14}\text{H}_{23}\text{N}^+(\text{CH}_3)$]. Similar observations have been seen in the literature (Khaled et al., 2003).

Adsorption Heat

An estimate of heat of adsorption was obtained from the trend of surface coverage with temperature as follows (Tang et al., 2006):

$$Q_{\text{ads}} = 2.303 R \left[\log \left(\frac{\theta_2}{1-\theta_2} \right) - \log \left(\frac{\theta_1}{1-\theta_1} \right) \right] \left(\frac{T_1 T_2}{T_2 - T_1} \right) \quad (10)$$

Where θ_1 and θ_2 are the degrees of surface coverage at temperatures T_1 and T_2 . The obtained value for $T_1 = 298$ K and $T_2 = 328$ K is $-81.2 \text{ kJ.mol}^{-1}$. The negative (Q_{ads}) value obtained in the case of the adsorption of HDTAB on the aluminium alloy (AA 3003) indicates a decrease of the degree of surface coverage with rise in temperature, supporting the earlier proposed physisorption mechanism (Oguzie et al., 2007).

Conclusion

From the overall experimental results and discussion the following conclusions can be deduced:

- i. Hexadecyltrimethylammonium bromide (HDTAB) acts as good inhibitor for the corrosion of aluminium alloy (3003) in 1 M HCl solution.
- ii. The inhibition efficiency increases with increase in inhibitor concentrations and the maximum value is 95.8 at 10^{-2} M for $T = 35^\circ\text{C}$.
- iii. The inhibition efficiency decreases with increase in temperature; the value of (%IE) is 97.2 at $T = 25^\circ\text{C}$ and 63.2 at $T = 55^\circ\text{C}$.
- iv. The adsorption of HDTAB on the aluminium alloy (AA 3003) surface obeys the modified Langmuir adsorption isotherm model.
- v. Phenomenon of physical adsorption is proposed from the thermodynamic adsorption and activation parameters obtained.

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