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Effects of ethanolamine and ethylamine on the entropy content of the corrosion of galvanised steel in 1mol/l NaCl solution

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ABSTRACT

The influence of ethanolamine and ethylamine on the corrosion behaviour of galvanised steel in 1mol/l NaCl solution was investigated at room temperature (25^{o}C) by weight loss method. Results obtained show that the compounds are better inhibitors at low concentrations (<0.5mol/l). From the decreasing weights of the metal over time, the concentrations of the metal was evaluated and plots of logarithm of the concentration of metal versus time as well as Erying-type equations facilitated the access of thermodynamic parameters such as equilibrium constant (k), enthalpy (“H*) and entropy (“S*) of the corrosion process. The entropy of the aggressive ions was found to have reduced on introduction of the inhibitors into the corrodent medium. Attempt to correlate the molecular structures of the inhibitors to the mechanism of inhibition was made.

KEYWORDS

Corrosion inhibition;
Galvanised steel;
Ethylamine;
Ethanolamine;
NaCl solution;
Entropy content.

INTRODUCTION

In recent years, more efforts have been directed on the investigation of metallic corrosion, probably due to the increase in the use of metals in engineering construction, technological related field and increase in environmental pollution in which one of its major effects is the corrosion of the metals exposed to the environment. The quest to reduce the extent to which corrosion has destroyed these structures seems to have drawn the attention of many. This is seen in the numerous literatures on the topic.

The use of corrosion inhibitors is one of the most practical methods for protection of metals against corrosion[1]. Most well known inhibitors are organic compounds containing nitrogen, sulphur, oxygen and phosphorus in their functional groups[2]. Adsorption of these compounds on the metal surface is the main mechanism of inhibition and it depends on the nature of the charge of the metal, the type of electrolyte (corrodent) medium, the chemical structure and reactivity of the inhibitor[3]. The inhibition efficiency has been found to depend on some physic-chemical and electronic properties of the organic inhibitor which relate to its functional groups, steric effects, electronic density of the donor atoms, stability of the formed chelate and strong coordination if any[1,4,9,10]. El Ouali et al.[5] have presented the adsorption process on the metal surface whereby water molecules are displaced from a corroding interface as follows:

$$\mathrm{Org}_{(sol)} + n\mathrm{H_2O}_{(ads)} \rightarrow \mathrm{Org}_{(ads)} + n\mathrm{H_2O}_{(sol)}$$

Hitherto, various kinetic and thermodynamic parameters have been monitored in corrosion of metals via weight loss trends, gasometric methods, thermometric methods, etc.[1,5-8]. This particular paper envisages the adsorption entropy content of the medium in which corrosion occurs at room temperature. From a classical-molecular conception, the rate of a chemical reaction is associated with the degree of availability of the particles of the reactants. When the molecules of the reactants are freer to move about and collide more effectively, the surface area of the reacting species is said to have increased and this in turn increases the reaction rate. In a system where the molecules are allowed to move freer, the system is said to be highly disordered. And this is the classical-molecular description of entropy. It is believed that the cumulative entropies of the species in the aggressive medium are responsible for the corrosion rate of the metal. While the inhibitive effect of the organic compounds is to protect the active centres on the metal surface thereby hindering attacks by the aggressive species and as a result decrease the adsorption entropy with respect to the reaction at the galvanized steel/aggressive species interface which in turn reduces corrosion rate.

EXPERIMENTAL

Materials

The sheets of commercially available galvanised steel used for this study were identified and obtained locally. Each sheet was 0.10cm in thickness. The sheets were mechanically press cut into 2.50cm by 2.50cm coupons. The galvanized steel coupons were polished to remove unwanted impurities using emery papers, degreased with acetone, washed in distilled water and air-dried before use[11,12]. The concentrations of the sodium chloride, ethanolamine and ethylamine solutions were prepared by dilution method[14,15]. They chemicals used were Analytical Grade (BDH) without further purification[6].

Weight loss measurements

The galvanized steel coupons were polished with emery papers to remove adhering impurities, degreased with acetone and air-dried. The specimens were immersed in five 100ml beakers (in two sets) and that of the blank (1mol/l NaCl). The first sets contained 1mol/l NaCl in each and a corresponding 0.2472mol/l, 0.4944mol/l, 0.7416mol/l, 0.9888mol/l and 1.2360mol/l of ethanolamine respectively. The second sets contained 0.2285mol/l, 0.4569mol/l, 0.9138mol/l and 1.1423mol/l ethylamine respectively. The inhibitors were prepared from the stock solution following a uniform trend of volumes corresponding to their calculated concentrations given above viz.: 1.5ml, 3.0ml, 4.5ml, 6.0ml and 7.5ml. The galvanized steel coupons were hanged into the experimental solution with the help of glass hooks. The initial weights of the specimens were noted. The variation in weight loss was monitored at 5days interval progressively for 25days. After every 5days the specimens were removed, polished with emery papers, washed in distilled water, degreased with acetone, air-dried and final weights noted. From the initial and final weights of the specimens, the loss of weights was calculated and the inhibitor efficiency (%IE) was calculated using equation (1a)[4, 6-8, 11, 16]:

$$\begin{array}{l}\textbf{Efficiency of inhibitors or}\\ \textbf{inhibition efficiency (\%IE)}\end{array} = \frac{W_0 - W_1}{W_0} \times 100 \qquad \textbf{(1a)}$$

where W_0 is the weight loss without inhibitor and W_1 is the weight loss with inhibitor.

The corrosion rate of galvanized steel was calculated (in mpy^{-1} –millimetre penetration per year) using the equation[1, 6-7]:

$$\mathrm{CR} = \frac{87.6\,W}{DAT} \qquad \textbf{(1b)}$$

where W =weight loss (g); D = density of galvanised steel (7.88gcm^{-3}); A = area of metal in cm$^{2;}$ T = exposure time (h).

RESULTS AND DISCUSSION

Weight loss measurements

The corrosion of galvanised steel in 1mol/l solution of NaCl with and without inhibitors were investigated at room temperature (25°C). The results obtained show that the corrosion rate increases as the period of immersion increases as shown in TABLE 1, 2 and 3. It has also been observed that the corrosion rates at higher concentration of the inhibitors are significantly large than that found at lower concentrations.

TABLE 1 : The results of corrosion of galvanized steel in 1mol/l NaCl with time

Time (Days)	Final Weight (g)	Conc. Fe,[Fe] (mol/l)	log [Fe] (mol/l)	CR (mpy^{-1})
0	4.558	0.814	-0.089	-
5	4.439	0.792	-0.101	1.7 x10^{-2}
10	4.351	0.777	-0.109	1.5 x10^{-2}
15	4.292	0.766	-0.116	1.2 x10^{-2}
20	4.235	0.756	-0.121	1.1 x10^{-2}
25	4.211	0.752	-0.123	1.0 x10^{-2}

Ethanolamine shows limiting corrosion inhibition efficiency at a concentration of 0.5mol/l. At concentration higher than 0.5mol/l, the compound behaves in a rather inconsistent manner and tends to promote corrosion. Ethylamine also inhibits better below 0.5mol/l but not as inefficient as ethanolamine above 0.5mol/l. This is shown in the trend of corrosion rates and inhibition efficiencies of the compounds as obtained in TABLE 2 and 3. However, it is of assumption that both compounds would be better inhibitors at lower con-

TABLE 2 : The results of corrosion of galvanized steel in 1mol/l NaCl with ethanolamine (ETA) as inhibitor

Vol. of Inhibitor in 100ml of 1mol/l NaCl (ml)	Conc. of ethanolamine (ETA), (mol/l)	Time (Days)	Final Weight of Fe (g)	Conc. Fe,[Fe] (mol/l)	log[Fe] (mol/dm^3)	CR(mpy^{-1})	%IE
1.5	0.2471	0	4.369	0.780	-0.108	-	-
		5	4.326	0.774	-0.111	5.5x10	68.87
		10	4.265	0.756	-0.122	7.3x10	52.63
		15	4.243	0.732	-0.135	6.5x10	50.24
		20	4.138	0.691	-0.161	8.6x10	28.48
		25	4.108	0.634	-0.198	9.4x10	8.60
3.0	0.4944	0	4.334	0.774	-0.111	-	-
		5	4.268	0.768	-0.118	9.7x10	44.74
		10	4.135	0.738	-0.132	9.9x10	36.13
		15	3.929	0.701	-0.154	1.0x10	35.89
		20	3.679	0.657	-0.182	9.3x10	22.60
		25	3.376	0.603	-0.220	1.0x10	12.68
4.5	0.7416	0	4.067	0.726	-0.139	-	-
		5	4.000	0.714	-0.146	9.9x10	44.01
		10	3.875	0.692	-0.160	9.3x10	40.01
		15	3.654	0.653	-0.185	1.1x10	16.80
		20	3.348	0.598	-0.223	1.3x10	5.26
		25	3.014	0.538	-0.269	1.0x10	3.58
6.0	0.9888	0	4.266	0.762	-0.118	-	-
		5	4.216	0.753	-0.123	7.4x10	58.27
		10	4.114	0.735	-0.134	7.6x10	51.20
		15	3.917	0.699	-0.155	9.7x10	26.05
		20	3.635	0.649	-0.188	1.0x10	12.69
		25	3.328	0.594	-0.226	1.0x10	11.53
7.5	1.2360	0	4.393	0.784	-0.105	-	-
		5	4.325	0.772	-0.112	1.0x10	42.58
		10	4.187	0.748	-0.126	1.0x10	34.21
		15	3.952	0.706	-0.151	1.2x10	11.76
		20	3.646	0.651	-0.186	1.1x10	5.26
		25	3.309	0.591	-0.228	1.0x10	2.88

TABLE 3 : The results of corrosion of galvanized steel in 1mol/l NaCl with ethylamine (EA) as inhibitor

Vol. of Inhibitor in 100ml of 1mol/l NaCl (ml)	Conc.of Ethylamine (EA), (mol/l)	Time (Days)	Final Weight of Fe (g)	Conc. Fe,[Fe] (mol/l)	log[Fe] (mol/l)	CR(mpy^{-1})	%IE
1.5	0.2285	0	4.346	0.776	-0.110	-	-
		5	4.329	0.773	-0.112	2.5x10	85.71
		10	4.299	0.767	-0.115	3.4x10	77.51
		15	4.285	0.765	-0.116	3.0x10	77.07
		20	4.224	0.754	-0.122	4.5x10	62.23
		25	4.171	0.745	-0.128	5.3x10	49.57
3.0	0.4569	0	4.507	0.805	-0.094	-	-
		5	4.471	0.798	-0.097	5.3x10	69.75
		10	4.416	0.789	-0.103	6.7x10	56.46
		15	4.388	0.784	-0.106	5.9x10	55.26
		20	4.286	0.765	-0.116	8.2x10	31.58
		25	4.219	0.753	-0.123	8.5x10	17.0
4.5	0.6854	0	4.152	0.741	-0.130	-	-
		5	4.120	0.736	-0.133	4.7x10	56.300
		10	3.988	0.712	-0.148	9.8x10	36.84
		15	3.811	0.681	-0.167	8.7x10	33.46
		20	3.594	0.642	-0.192	8.0x10	32.82
		25	3.302	0.570	-0.229	8.7x10	15.85
6.0	0.9138	0	4.600	0.821	-0.085	-	-
		5	4.550	0.813	-0.090	7.4x10	50.24
		10	4.452	0.795	-0.100	7.2x10	47.06
		15	4.295	0.767	-0.115	7.8x10	44.36
		20	4.090	0.730	-0.136	7.6x10	35.16
		25	3.840	0.686	-0.164	8.7x10	33.75
7.5	1.1423	0	4.537	0.810	-0.091	-	-
		5	4.478	0.800	-0.097	8.7x10	50.74
		10	4.355	0.778	-0.109	9.1x10	41.63
		15	4.183	0.747	-0.127	8.5x10	35.16
		20	3.969	0.709	-0.150	7.9x10	33.75
		25	3.696	0.660	-0.180	8.1x10	21.21

centrations in the corrodent. Figure 1a and 1b shows that ethylamine at both small and high concentrations (volumes) inhibits better than ethanolamine.

Application of absolute reaction rate theory

The theory of absolute reaction rates[17], frequently also called the transition-state theory, is based on statistical mechanics and represents an alternative approach to reaction kinetics. This Theory postulates that molecules before undergoing reaction must form an activated complex in equilibrium with the reactants, and that the rate of any reaction is given by the rate of decomposition of the complex to form the reaction products.

For a reaction between a molecule of A and one of B, the postulated steps can be represented by the scheme

$$\mathbf{A + B \rightleftharpoons [A.B]^* \xrightarrow{K} Products}$$

reactants **activated complex**

The activated complex has certain properties of an or-

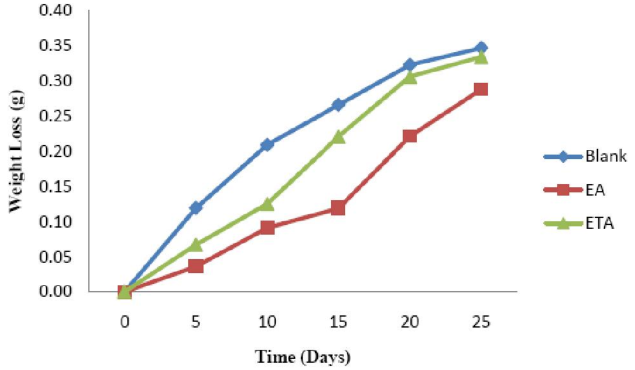


Figure 1a : Variation of weight loss of galvanized steel without and with 3ml of inhibitors (EA and ETA) in 1mol/l NaCl solution

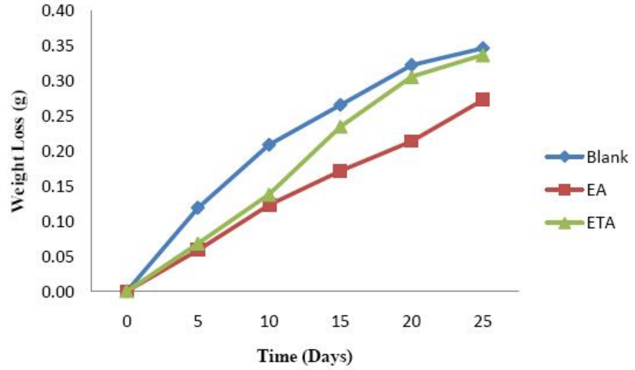


Figure 1b : Variation of weight loss of galvanized steel without and with 7.5ml of inhibitors (EA and ETA) in 1mol/l NaCl solution

dinary molecule and possesses temporary stability.

Following the above theory, the corrosion mechanism is postulated with the scheme

$$\mathbf{Fe_{(s)} + NaCl_{(aq)} \underset{}{\overset{K^*}{\rightleftharpoons}} [Fe(H_2O)_6]Cl_3}$$

$$\mathbf{\xrightarrow{*K} Fe2O3{\cdot}xH_2O + FeCl_3} \qquad (2)$$

Following the above ideas, Eyring[17] showed that the rate constant k of any reaction irrespective of the order or molecularity is given by the expression

$$\mathbf{k = \frac{RT}{Nh} K*} \qquad (3)$$

where R is the gas constant; N, Avogadro’s number; h, Plank’s constant; T, the absolute temperature; and K*, the equilibrium constant for the reaction of the activated complex from the reactants.

We resort to thermodynamics and write for K*

$$\mathbf{InK* = \frac{-\Delta G*}{RT}} \qquad (4)$$

$$\mathbf{InK* = \frac{(\Delta H* - \Delta TS*)}{RT}} \qquad (5)$$

where “G*, “H* and “S* are respectively the free energy, enthalpy and entropy of activation.

Introducing equation (5) into equation (3) we obtain for k

$$\mathbf{k = \frac{RT}{Nh} exp\left(\frac{\Delta S*}{R}\right) exp\left(\frac{-\Delta H*}{RT}\right)} \qquad (6)$$

$$\mathbf{Ink = In\left(\frac{RT}{Nh}\right) + \frac{\Delta S*}{R} - \frac{\Delta H*}{RT}} \qquad (7)$$

Consequently when k and “H* of a reaction are known at a given temperature, “S* may be found.

$R = 8.314JK^{-1}mol^{-1}$; $N = 6.023 \times 10^{23}mol^{-1}$; $h = 6.626 \times 10^{-34}Js$

$$\mathbf{\therefore Slope = \frac{-k}{2.303}}$$

The equilibrium constant (k) of the reaction in the Eyring-type equation (eqn.7) has been extrapolated from the slope of the straight line graph of the natural logarithm of the concentration of galvanised steel (calculated from molar mass-weight of substance relation) and time as shown in Figure 2 and 3 and TABLE 4.

From equation 2, the enthalpy of the reaction was evaluated from standard tables to be 816.8J/K/mol since the reaction was carried out at constant room temperature. Hence, the entropy of the reaction at different concentrations of the inhibitors was calculated from k and “H*.

It can be seen from TABLE 4 that the values of

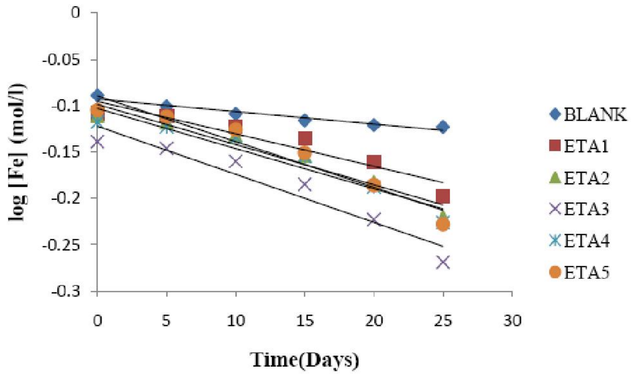


Figure 2 : Plot of log [Fe] (mol/l) versus time (days) for galvanized steel corrosion in 1mol/l NaCl(Blank) with 1.5ml(ETA1), 3.0ml(ETA2), 4.5ml(ETA3), 6.0ml(ETA4) and 7.5ml(ETA5) of ethanolamine as inhibitor at room temperature(273.15K)

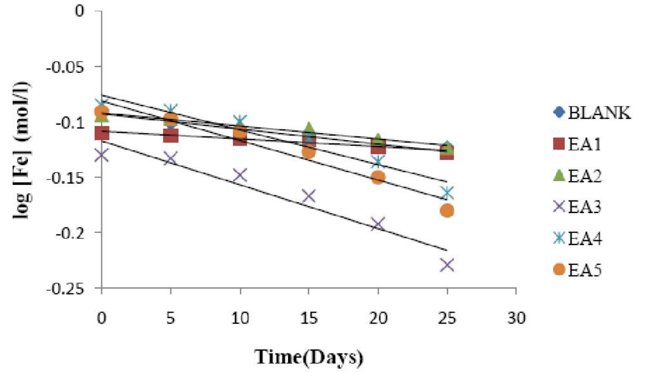


Figure 3 : Plot of log [Fe] (mol/l) versus time (days) for galvanized steel corrosion in 1M NaCl(Blank) with 1.5ml(EA1), 3.0ml(EA2), 4.5ml(EA3), 6.0ml(EA4) and 7.5ml(EA5) of ethylamine as inhibitor at room temperature(273.15K)

entropy (ΔS^*) in the presence of inhibitors are negative and larger than in the absent of inhibitors. This means that the aggressive ions were in higher order state than at the initial state[1]. The decrease in the solvent entropy is as a result of desorption of water/aggressive ions that were adsorbed on the surface of the metal which were followed by adsorption of the inhibitors on the surface of the metal[18].

The solution which had the aggressive ions in the most ordered state was obtained with about 0.2mol/l of ethylamine as inhibitor with ΔS^* value of -301.16K/J/mol with a corresponding highest corrosion efficiency of 85.71%. This excellent inhibitory effect merits to be investigated at similar or lower concentrations and varying temperatures using other conventional techniques of corrosion studies.

TABLE 4 : Thermodynamic parameters for the corrosion of galvanized steel in 1mol/l NaCl with and without the presence of different concentrations of inhibitors at room temperature (298K)

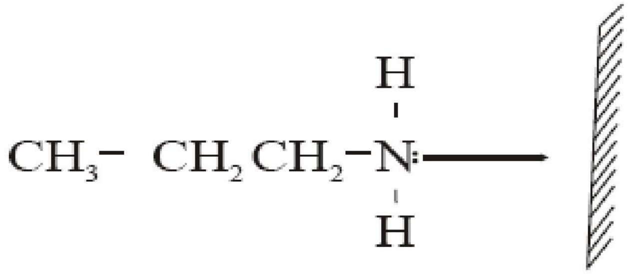
System	Concentration (mol/l)	K	ΔH^* (J/K/mol)	ΔS^* (J/K/mol)
Blank	1.00	0.004	-816.30	-274.41
Ethanolamine HO-CH_2-CH_2-NH_2	0.2471	0.007		-288.89
	0.4944	0.009		-286.80
	0.7416	0.012		-284.41
	0.9888	0.099		-266.86
	1.2360	0.115		-265.61
Ethylamine CH_3-CH_2-NH_2	0.2285	0.002		-301.16
	0.4569	0.003		-295.93
	0.6854	0.006		-286.80
	0.9854	0.007		-288.89
	1.1423	0.008		-287.78

Mechanism of corrosion inhibition

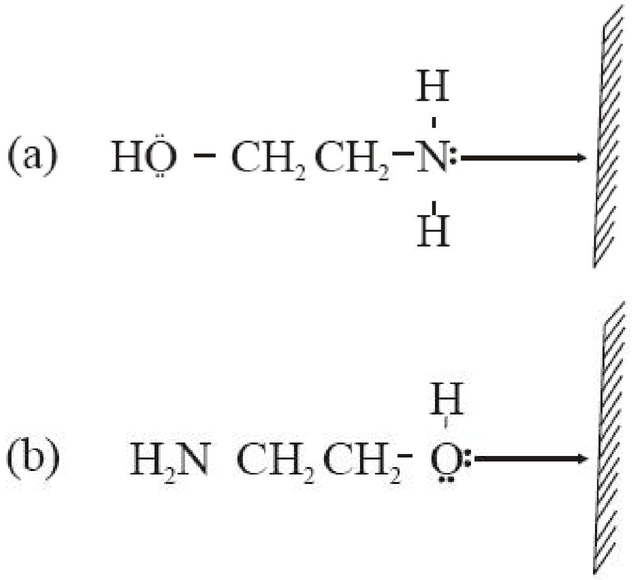
The inhibition of corrosion can be explained on the basis of the concept of adsorption of inhibitors on the corroding metal surface[16]. The inhibitive action of these compounds has been attributed to the strong adsorption of these molecules on the metal surface using the lone pairs of electron available on the heteroatoms.

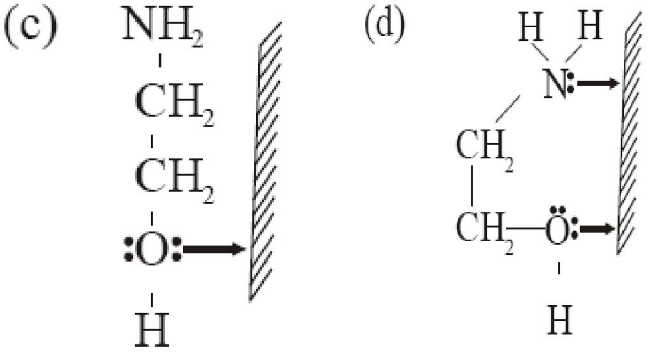
The effective inhibition by ethylamine is because the molecule has less interaction with the medium as the nitrogen lone pairs of electron anchors on the positive metal surface. While ethanolamine on the other hand, though with more lone pairs of electron has the OH group which at high concentration has been suspected to be responsible for the gradual deterioration of the metal. The corrosion inhibition mechanism of the compounds is depicted in Scheme 1 and 2:

In Scheme 2 (a), it has been shown that there is a high inductive effect towards the more electronegative



Scheme 1





Scheme 2

hydroxyl group thereby causing a possible desorption of the adsorption centre, the amine group from the metal surface. Thus, there would have been similar effect from the two inhibitors as observed from the results if there was only the amine group available for adsorption.

From Scheme 2 (b), the attack of the metal surface by the hydroxyl group may have resulted in the formation of a soluble metal oxide, hence increase in corrosion rate compared to the action of ethylamine. The inhibitive action of ethanolamine is related to the joint action of the amine and hydroxyl groups. While the amine group does the inhibition, the hydroxyl group promotes corrosion. This has reflected in the results obtained for the weight loss of galvanized steel in 1M NaCl using ethanolamine as inhibitor as shown in TABLE 2.

Scheme 1 (c) and (d) shows another possible adsorption modes of the inhibitors based on the orientation of the functional groups in ethanolamine during an attack. If scheme 2 (c) occurred, then there must have been a possible steric hindrance on the metal surface for the adsorption of other molecules available in the solution and possibly the effective adsorption of the nitrogen lone pairs of electron on the metal surface may be impaired. This could lead to physical adsorption of the other part of the molecules on the metal surface thereby reducing corrosion while the hydroxyl group which has a negative effect attacks the other part of the metal surface. The probability for scheme 2 (d) to occur, though slim, due to the possible strain on the cyclic chain formed produces similar effect as in scheme 2 (c). Hence, there is possibility of occlusion of some part of the metal surface, while the both active centres are attacking independently.

Therefore, based on the results in TABLE 2 obtained for the corrosion inhibition of galvanized steel in 1mol/l NaCl by ethanolamine, it is suspected that the amine group is responsible for the effective corrosion inhibition while the hydroxyl group is responsible for the deterioration of the metal. This result is synonymous with the joint action of the two groups in scheme 2 (d) and possibly scheme 2 (c) or combined action of scheme 2 (a) and 2 (b) respectively.

CONCLUSION

Galvanized steel exhibits good resistance to corrosion in saline medium until the anodically protected surface is penetrated.

The two inhibitors used in this study -ethanolamine and ethylamine provided a good inhibition effect but are not good inhibitors at higher concentrations in the corrodent medium considered. Ethylamine has greater corrosion inhibition efficiency than ethanolamine in the corrodent medium considered.

The entropy of the aggressive ions was found to have reduced on introduction of the inhibitors into the corrodent medium. Consequently raising the activation energy of the corrosion process.

The effectiveness of the inhibitors clearly signifies the action of their molecular structures.

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