



Effect of Chloride Ions on the Corrosion Behavior of Al – Zn Alloy in NaOH Solution at Four Different Temperatures

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Abstract

This research involves study effect of chloride ions in concentration range ($0.01 - 0.50 \text{ mol.dm}^{-3}$) on the corrosion behavior of Al-Zn alloy in basic media of $1 \times 10^{-3} \text{ mol.dm}^{-3}$ NaOH at pH=11 and four different temperatures in the range (298-313 K). Cathodic and anodic Tafel slopes (b_c & b_a) and transfer coefficients (α_c & α_a) were calculated and the results interprets according to the variation of the rate – determining steps. The results also indicate that the chloride ions are bonded chemically in the interface as an initial step of formation of different mixed oxohydroxy – and chloro complexes. Polarization resistance (R_p) is calculates and interprets the different polarization behavior because of addition of chloride ions to the basic media.

Keywords: Al-Zn alloy, Effect of chloride ion, Corrosion behavior in basic medium.

1. Introduction:

Corrosion of aluminum has been a subject of numerous studies due to the importance of this material in contemporary civilization. It is well known that there is a potential region in which the rate of corrosion, even in such aggressive media as chlorides. The relatively complex corrosion mechanism of aluminium has been studies by several authors.

Corrosion of aluminium occurs only when the metal protective oxide layer is damaged and when the repair mechanism is prevented by chemical dissolution[1].

Polarization methods have been extensively used to investigate the mechanism of localized corrosion and processes that lead to localized corrosion. In using potentiostatic techniques, the potential variable[2]. Potentiostatic and potentiodynamic techniques have been applied by several authors to study the corrosion of aluminium in different environment[3-8].

The exceptional corrosion resistance of aluminium in many environments is due to its protective oxide film which is relatively inert chemically and so provides the passive behavior of aluminium.

However in aggressive environments, particularly in the presence of halide ions, aluminium suffers from localized corrosion by local breakdown of the passive film. Various theories are postulated to explain the mechanism of passivity breakdown in halide solutions but the most common and accepted ones are as follows:

- a- Penetration theory.
- b- Flaws and crack/heal theory.
- c- Localized acidification theory.
- d- Complex formation theory.

2. Experimental Work:

Al – Zn alloy was cut into cylinder shape with (1.2 cm) diameter, and made into electrode by pressing a copper wire into a hole on one side and then insulating all but one side with an epoxy resin. The open side was polished mechanically to a mirror finish, rinsed in distilled water and stored in a desiccator.

The analytical composition of alloy was shown in Table (1) which was obtained by chemical analysis in Naser Company.

Table 1
The Analytical Compositions of Al-Zn Alloy Which Useful in This Search.

Metal	Zn	Mg	Cu	Cr	Ni	Ti	Mn	Al
Wt %	5.70	2.50	1.330	0.22	0.01	0.08	0.06	Bal.

The electrochemical glass cell was of the usual type with provision for working electrode (Al-Zn alloy), auxiliary electrode (Pt electrode), and a Luggin capillary for connection with an saturated calomel electrode (reference electrode SCE). The basic solution was 1×10^{-3} mol.dm⁻³ NaOH (obtained by Ferak with M.wt 40 g.mol⁻¹ and purity >99.5%) and distilled water (specific conductivity 1×10^{-6} S.m⁻¹). To study the effect of chloride ions uses NaCl (obtained by Fluka with M.wt 58.44 g.mol⁻¹ and purity 99.5%) was prepared with seven concentrations (0.01, 0.10, 0.15, 0.20, 0.25, 0.35, and 0.50 mol.dm⁻³).

Electrochemical measurements were performed with a potentiostat (Corroscript) which was obtained from Tacussel (France) at a scan rate of 0.3 Volt per minute.

The Tafel extrapolation and Linear-polarization techniques are uses to determine the rate of corrosion. The Tafel extrapolation method for determining corrosion rate was used by Wagner and Traud to verify the mixed-potential theory, the method was shown in figure (1). Tangents to the anodic and cathodic Tafel regions were extrapolated to the point of intersection, from which both the corrosion potentials (E_{corr}) and corrosion current density (i_{corr}) were determine using the four-point method [9] cathodic (bc) and anodic (ba) Tafel slopes, transfer coefficients (α), polarization resistances (R_p) together with other data could be derived from the polarization curves by using extrapolation method.

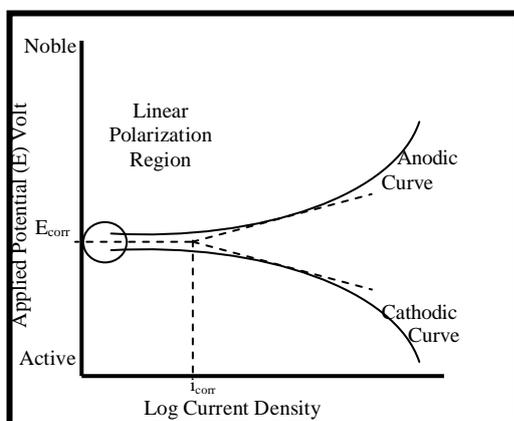
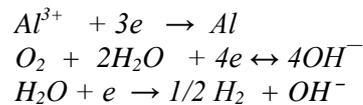


Fig.1. Tafel Extrapolation Method.

3. Results and Discussion:

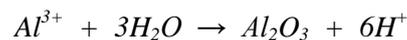
Figure (2) represent the polarization curve of Al – Zn alloy in 1×10^{-3} NaOH solution (pH=11) in the absence of chloride ions (Cl^-) at 298 K, the section (abc) represented the cathodic polarization region which occur on the metals presumably by migration of electrons through the surface oxide films and subsequent interaction of those electrons with water molecules and dissolved oxygen at the film/solution interface, so the corrosion process is complicated with mass transfer. Corrosion of Al-Zn alloy proceeds similarly by reduction of oxygen and the water molecules will act as the electron acceptor and the rate of corrosion is controlled by the diffusion limited current density for cathodic reduction, the probable reactions are:



Thus metals like aluminium and zinc, whose oxides are amphoteric, are thermodynamically unstable in alkaline solutions and will react with water at high pHs with consequent hydrogen evolution and formation of metal anions as shown below:



Along the section (cde), the metal hydroxide is expected to be formed. The hydroxide soon dissociates into metal oxide (Al_2O_3) on a surface which behaves as a passive layer (protective film) according to the reaction:



The breakdown of passivity began at point (e) and continued along (ef) and this section represent the anodic Tafel region.

The presence of Cl^- effects mainly on anodic polarization. Fig. (3) show the effect addition of $0.01 \text{ mol.dm}^{-3} Cl^-$ on the corrosion behavior, since addition Cl^- with this concentration penetrates the passive layer but partially destroyed and shifts passive potential (E_{pass}) to more negative values and corrosion current density to lower values.

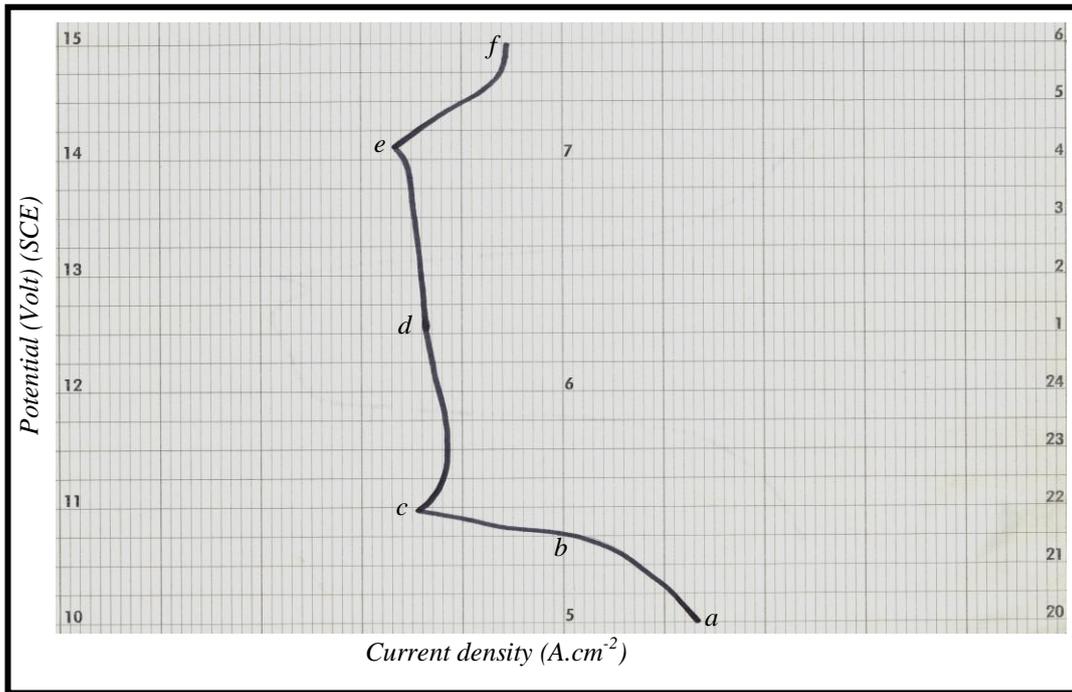


Fig.2. Polarization Curve for Corrosion of Al-Zn alloy in 1×10^{-3} mol.dm⁻³ NaOH Solution in the Absence of Chloride Ion at 298K.

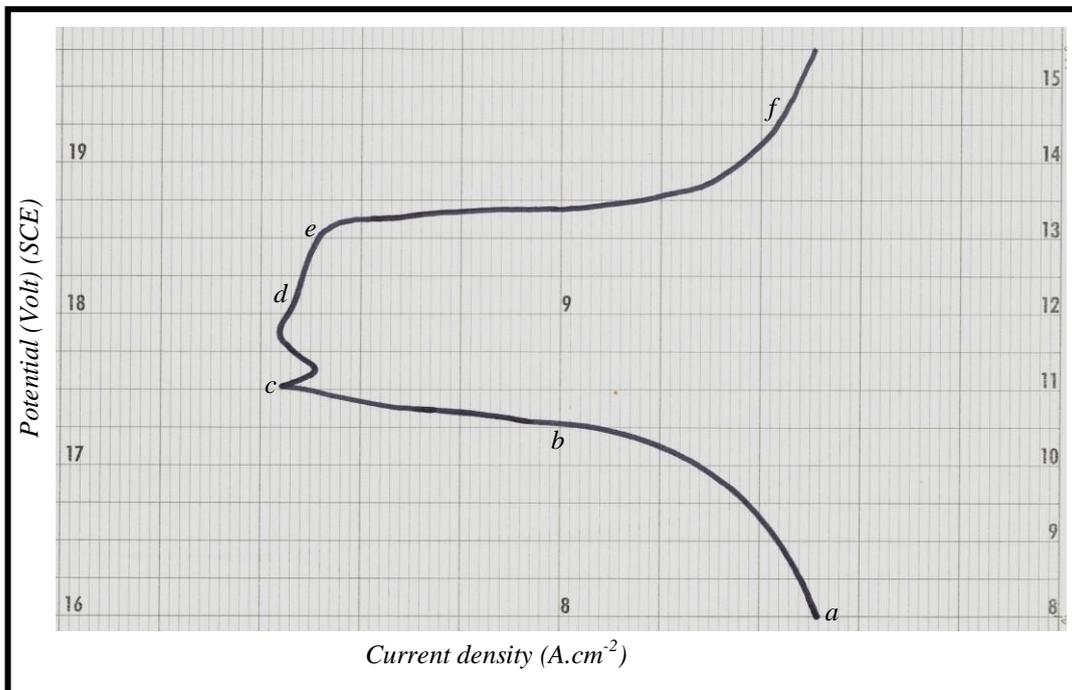


Fig.3. Polarization Curve for Corrosion of Al-Zn Alloy in 1×10^{-3} mol.dm⁻³ NaOH Solution in the Presence of 0.01 mol.dm⁻³ Chloride Ion at 298K.

When add a higher concentration of chloride ion, the passive layer was breakdown and (i_{corr}) value increases with increasing the concentration

of Cl^- in solution as shown in Figs.(4) to (9) where the polarization curve, in general, consist of the cathodic (abc) and anodic (cde) Tafel regions.

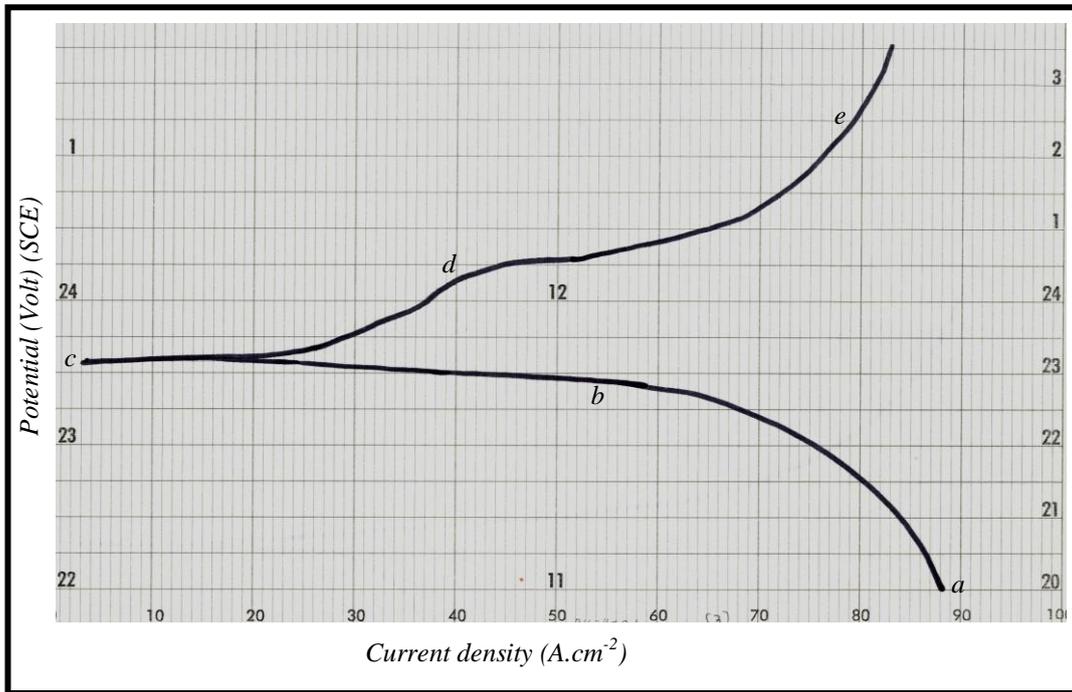


Fig.4. Polarization Curve for Corrosion of Al-Zn Alloy in 1×10^{-3} mol.dm⁻³ NaOH Solution in the Presence of 0.10 mol.dm⁻³ Chloride Ion at 298K.

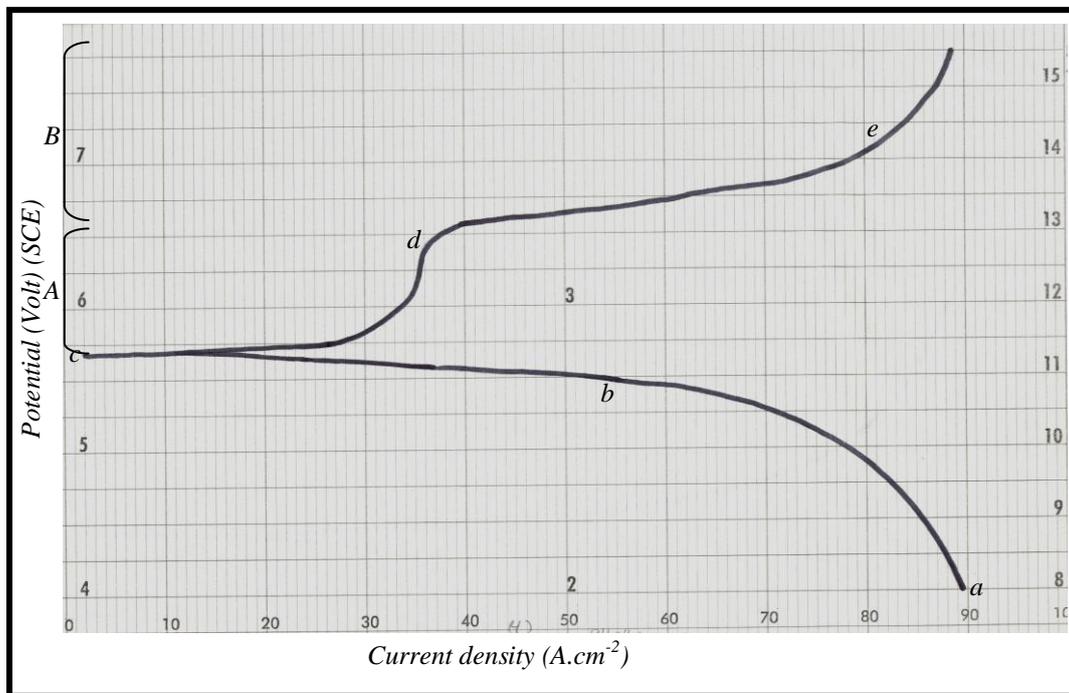


Fig.5. Polarization Curve for Corrosion of Al-Zn Alloy in 1×10^{-3} mol.dm⁻³ NaOH Solution in the Presence of 0.15 mol.dm⁻³ Chloride Ion at 298K.

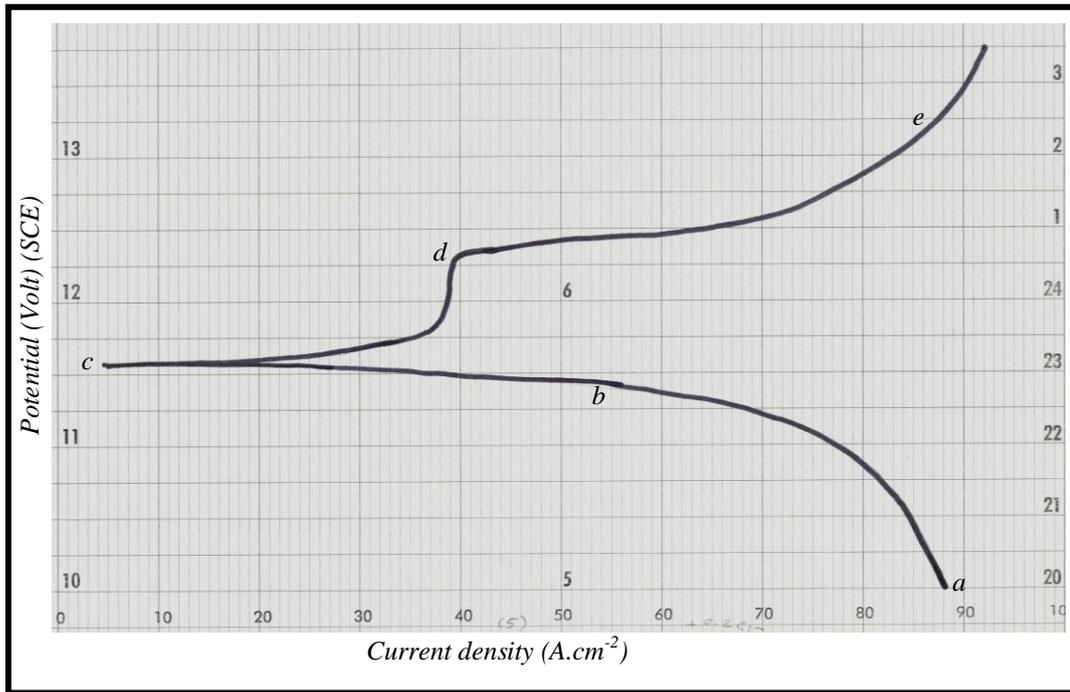


Fig.6. Polarization Curve for Corrosion of Al-Zn Alloy in 1×10^{-3} mol.dm⁻³ NaOH Solution in the Presence of 0.20 mol.dm⁻³ Chloride Ion at 298K.

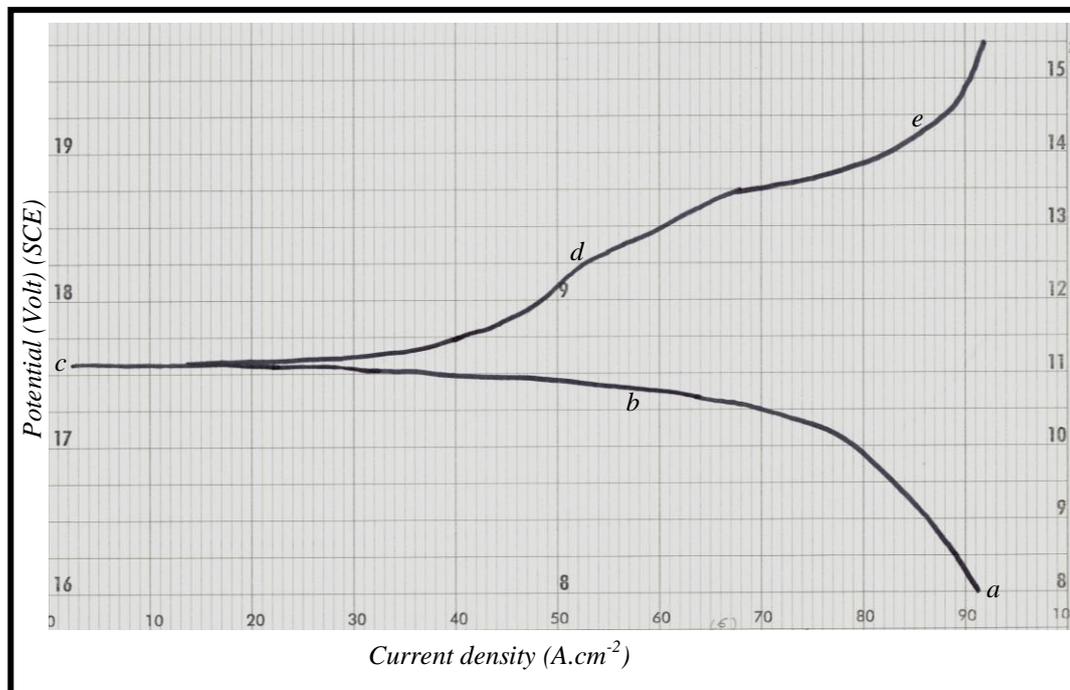


Fig.7. Polarization Curve for Corrosion of Al-Zn Alloy in 1×10^{-3} mol.dm⁻³ NaOH Solution in the Presence of 0.25 mol.dm⁻³ Chloride Ion at 298K.

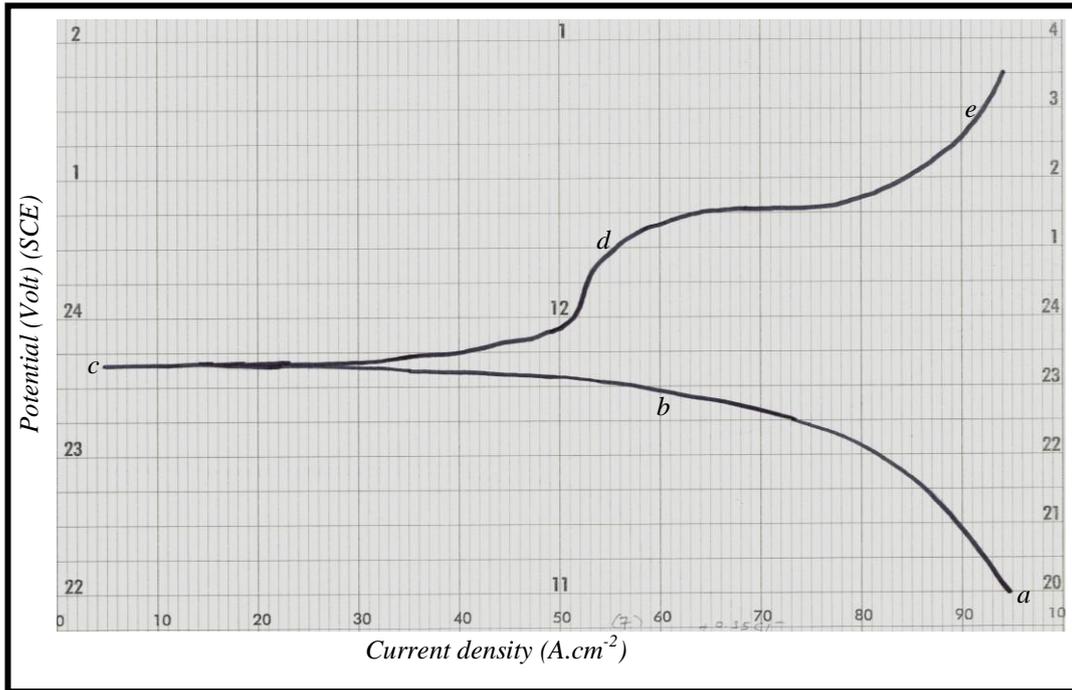


Fig.8. Polarization Curve for Corrosion of Al-Zn Alloy in 1×10^{-3} mol.dm⁻³ NaOH Solution in the Presence of 0.35 mol.dm⁻³ Chloride Ion at 298K.

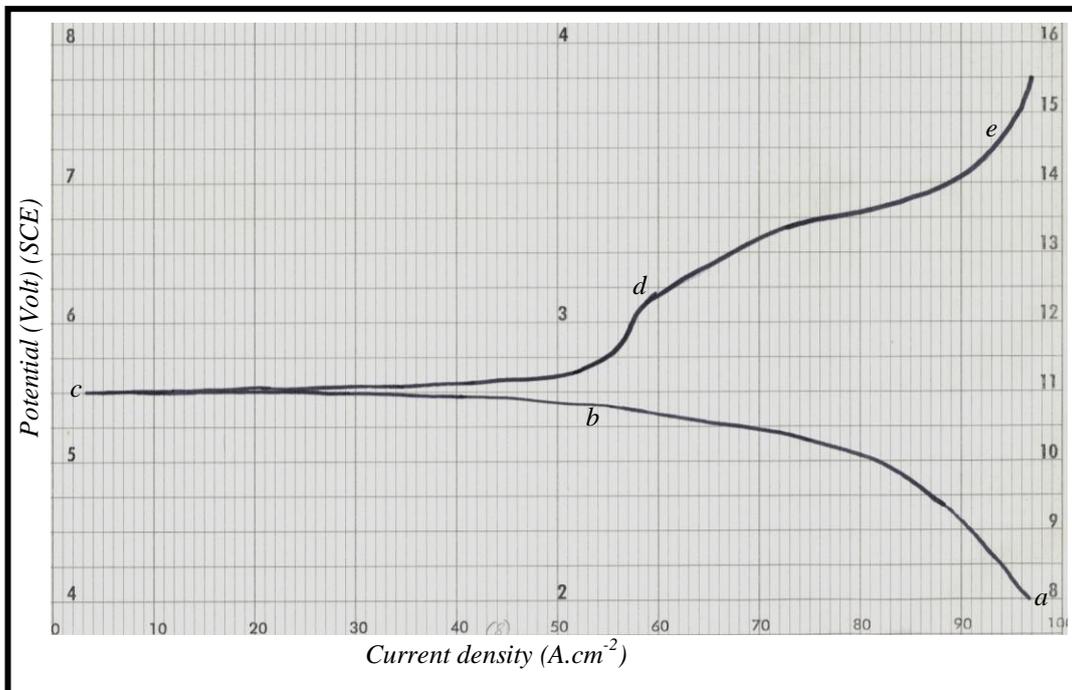
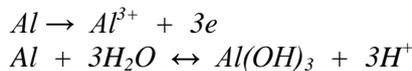


Fig.9. Polarization Curve for Corrosion of Al-Zn Alloy in 1×10^{-3} mol.dm⁻³ NaOH Solution in the Presence of 0.50 mol.dm⁻³ Chloride Ion at 298K.

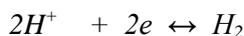
When only hydroxide ions (OH^-) exists in solution (NaOH without additive), aluminium hydroxide $\text{Al}(\text{OH})_3$ will be formed and so repairing the passive film on the anode.

Moreover the electrolyte at the anode will be replenished again with (OH^-) by migration. But when Cl^- exists in solution (NaOH with additive), then part of the current will be transported by these ions and since the anode products in alkaline chloride solution is still $\text{Al}(\text{OH})_3$, therefore the concentration of chloride ions at the anode will be increased rapidly while the concentration of hydroxide ions decreases to values corresponding to pH values less than (7), solid $\text{Al}(\text{OH})_3$ is still formed, but finally the electrolyte becomes so acid that it leads to the formation of soluble anode products rather than solids.

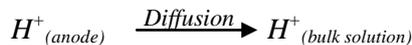
This process leads to the breakdown of passivity, the reactions may be given by:



Under equilibrium condition the rates of production and consuming of protons will be equal. The proton can be consumed by direct reduction:



Or by diffusion to the bulk solution:



The results of E_{corr} , i_{corr} , b_c , b_a , α_c , α_a , R_p , and i_0 for Al – Zn alloy in the absence and presence of chloride ions in NaOH solution at four different temperatures have been given in Table (2).

From deep analysis of the cathodic and anodic regions of the polarization curves, it was possible to derive data concerning:

- The cathodic (b_c) and anodic (b_a) Tafel slopes.
- The cathodic (α_c) and anodic (α_a) transfer coefficients. Values of (α) have been calculated from the corresponding values of the Tafel slope (b) using the relation [10,11]:

$$\alpha = \frac{2.303RT}{bF} \quad \dots(1)$$

where:

R : gas constant ($8.314 \text{ J.mol}^{-1}.\text{K}^{-1}$).

T : temperature in Kelvin scale.

F : Faraday constant (96500).

- The polarization resistance (R_p) may be defined as [12,13, 14]:

$$R_p = \frac{d(E - E_{\text{corr}})}{di} \quad \dots(2)$$

where E and E_{corr} are the applied corrosion potential (Volt) respectively, I is the current density (A.cm^{-2}).

For small polarization, one may write approximation [13,14]:

$$R_p = \frac{d(E - E_{\text{corr}})}{di} = \frac{E_{\text{corr}}}{i_{\text{corr}}} \quad \dots(3)$$

where E_{corr} and i_{corr} are the corrosion potential (V) and corrosion current density (A.cm^{-2}). The ratio ($E_{\text{corr}}/i_{\text{corr}}$) thus corresponds to the resistance of the metal/solution interface to charge-transfer reaction. It is also a measure of the resistance of the metal to corrosion in the solution in which the metal is immersed.

For low-field polarization [15]:

$$\frac{\eta}{i} = \frac{RT}{Fi_0} \quad \dots(4)$$

and

$$\frac{E_{\text{corr}}}{i_{\text{corr}}} = \frac{RT}{Fi_0} = R_p \quad \dots(5)$$

where $\eta = E - E_{\text{corr}}$ and i_0 is the equilibrium exchange current density (A.cm^{-2}). The reaction resistance (R_p), which mainly depends upon the equilibrium exchange current density (i_0) determines what may be termed the polarizability, i.e., what overpotential ($\eta = E - E_{\text{corr}}$) a particular current density needs or produces since:

$$R_p = \frac{d\eta}{di} = \frac{d(E - E_{\text{corr}})}{di} = \frac{E_{\text{corr}}}{i_{\text{corr}}} = \frac{RT}{Fi_0} \quad \dots(6)$$

The polarization resistance (R_p) was also determined in another way from Stern- Geary equation [16,17,18], where:

$$R_p = \left(\frac{dE}{di} \right)_{i=0} = \frac{b_a b_c}{2.303(b_a - b_c) i_{\text{corr}}} \quad \dots(7)$$

The values of R_p have been calculated from equation (7), which is presented in table (2).

The results of Table (2) indicates that the lowest value of ($b = 0.026$ or $\alpha = 2.4$) and the highest value is ($b = 0.120$ or $\alpha = 0.5$). A cathodic Tafel slope of 0.120 (or $\alpha = 0.5$) may be diagnostic of discharge-chemical desorption mechanism for hydrogen evolution reaction of the cathode in which the proton discharge is the rate-determining

step. If chemical desorption is the rate-determining step, the rate will then be independent on the overpotential since no charge transfer occur in such step and the rate becomes directly proportional to the concentration or the coverage (θ) of the adsorbed hydrogen atoms, and may occur at coverage ranging from very small values to almost full surface layer formation [19].

Table 2

Values of Corrosion Potential (E_{corr}), Corrosion Current Density (i_{corr}), Cathodic and Anodic Tafel Slopes (b_c & b_a), Cathodic and Anodic Transfer Coefficients (α_c & α_a), Polarization Resistance (R_p), and Equilibrium Exchange Current Density (i_0) for Corrosion of Al-Zn Alloy in 1×10^{-3} mol.dm⁻³ NaOH Solution in Absence and Presence of Chloride Ions at Four Different Temperatures.

Medium (mol.dm ⁻³)	T (K)	Corrosion		b (V.decade ⁻¹)		α		Passivity		Rp/10 ⁺⁴	i ₀ /10 ⁻⁶ (A.cm ⁻²)
		-E _{corr} (Volt)	i _{corr} /10 ⁻⁶ (A.cm ⁻²)	-b _c	+b _a	α_c	α_a	-E _{pass} (Volt)	i _{pass} /10 ⁻³ (A.cm ⁻²)		
1x10 ⁻³ NaOH (pH=11) only	298	1.66	2.916	0.107	-	0.55	-	0.99	0.501	14.910	0.172
	303	1.61	3.125	0.100	-	0.60	-	1.20	0.542	13.913	0.187
	308	1.56	3.333	0.095	-	0.64	-	1.22	0.581	13.044	0.203
	313	1.50	3.750	0.091	-	0.68	-	1.24	0.622	11.594	0.232
pH=11 + 0.01 Cl ⁻	298	1.44	0.416	0.061	-	0.97	-	1.21	0.063	104.52	0.024
	303	1.39	0.500	0.058	-	1.04	-	1.24	0.071	86.956	0.034
	308	1.36	0.541	0.052	-	1.17	-	1.28	0.088	80.366	0.033
	313	1.33	0.625	0.042	-	1.45	-	1.30	0.010	69.565	0.002
pH=11 + 0.10 Cl ⁻	298	1.39	0.541	0.044	0.120	1.34	0.41	-	-	2.584	0.993
	303	1.36	0.583	0.034	0.111	1.76	0.55	-	-	1.938	1.346
	308	1.33	0.625	0.031	0.081	1.97	0.75	-	-	1.557	1.703
	313	1.31	0.666	0.029	0.062	2.14	1.00	-	-	1.288	2.093
pH=11 + 0.15 Cl ⁻	298	1.38	0.541	0.054	0.115	1.09	0.51	-	-	2.949	0.870
	303	1.33	0.583	0.047	0.100	1.27	0.60	-	-	2.381	1.096
	308	1.32	0.708	0.029	0.071	2.11	0.86	-	-	1.262	2.101
	313	1.31	0.958	0.026	0.051	2.38	1.21	-	-	0.780	3.454
pH=11 + 0.20 Cl ⁻	298	1.39	0.666	0.060	0.120	0.98	0.49	-	-	2.607	0.984
	303	1.37	0.833	0.049	0.093	1.22	0.64	-	-	1.672	1.560
	308	1.34	0.916	0.042	0.083	1.45	0.73	-	-	1.321	2.007
	313	1.32	1.416	0.027	0.068	2.29	0.91	-	-	0.592	4.550
pH=11 + 0.25 Cl ⁻	298	1.39	1.458	0.055	0.100	1.07	0.59	-	-	1.056	2.429
	303	1.38	2.000	0.049	0.096	1.22	0.62	-	-	0.704	3.706
	308	1.34	2.083	0.045	0.049	1.35	1.24	-	-	0.488	5.426
	313	1.33	2.500	0.031	0.036	2.00	1.72	-	-	0.289	9.321
pH=11 + 0.35 Cl ⁻	298	1.38	1.666	0.052	0.057	1.13	1.03	-	-	0.708	3.622
	303	1.35	1.875	0.047	0.055	1.27	1.09	-	-	0.586	4.447
	308	1.33	3.333	0.046	0.050	1.32	1.22	-	-	0.312	8.501
	313	1.32	3.750	0.042	0.042	1.47	1.47	-	-	0.243	11.09
pH=11 + 0.50 Cl ⁻	298	1.39	1.875	0.065	0.091	0.91	0.65	-	-	0.878	2.923
	303	1.38	2.242	0.058	0.081	1.03	0.74	-	-	0.654	2.988
	308	1.37	3.513	0.051	0.058	1.19	1.05	-	-	0.335	7.911
	313	1.36	4.445	0.043	0.037	1.44	1.67	-	-	0.194	13.88

The expected Tafel slope in such step would then be $-0.03\text{V.decade}^{-1}$ (or $\alpha=2.0$).

When electrochemical desorption becomes the rate-determining step for the hydrogen evolution reaction on the cathode, the expected value of b_c is $-0.05\text{V.decade}^{-1}$ (or $\alpha=1.5$).

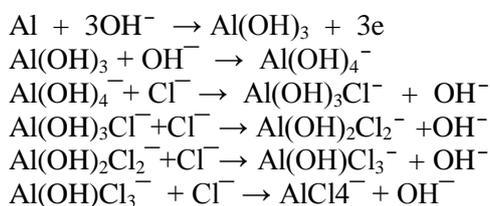
The variation of the cathodic and anodic Tafel slopes or transfer coefficients may be attributed to variation of the rate-determining step in the metals dissolution reaction.

Another approach to the problem of the electrochemical corrosion rate measurement is to apply only a small potential difference to the specimen and then measure the current density.

The measurement of polarization resistance has very similar requirements to the measurement of full polarization curves and it is particularly useful as a method to rapid identifying corrosion, up-setting and initiating a remedial action[20].

The results of Table (2) indicates that the values of R_p which obtained for the solution at $\text{pH}=11$ and in the ($\text{pH}=11+0.01 \text{ mol.dm}^{-3} \text{ Cl}^-$) system were higher than other media.

Generally, the polarization resistance (R_p) in the other media decreases with increasing the concentration of chloride ion in solution because of increases the attack on the bare surface. The higher values of R_p enhanced by presence the passive layer as shown in the polarization curves which attribute to produce aluminium hydroxide $\text{Al}(\text{OH})_3$ which transform to protective film (Al_2O_3) and reduces the expected attack on the surface alloy and lead to more resistance to corrosion as shown in Fig.(2), and to the equilibrium states between Al_2O_3 and AlCl_3 in addition to form $\text{Al}(\text{OH})_3$ in ($1 \times 10^{-3} \text{ NaOH} + 0.01 \text{ Cl}^-$) system as shown in Fig.(3), but when add higher concentrations of Cl^- there are many soluble complexes to be form according to the following reactions:

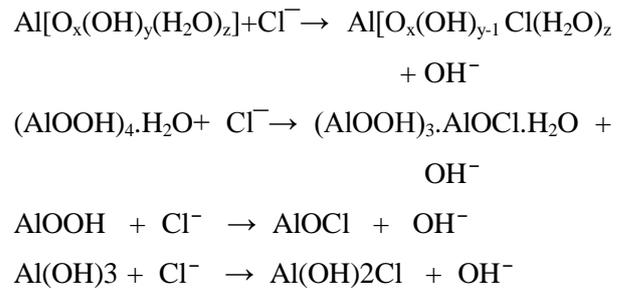


4. Conclusion:

From the present measurements the conclusion indicates that the polarization behavior of Al-Zn alloy in $1 \times 10^{-3} \text{ mol.dm}^{-3} \text{ NaOH}$ solution ($\text{pH}=11$) gives special case in the presence of $1 \times 10^{-2} \text{ mol.dm}^{-3} \text{ Cl}^-$ and this phenomena may be

attributed to occur the equilibrium state between the Al_2O_3 and AlCl_3 in addition to produce $\text{Al}(\text{OH})_3$, while the addition of higher concentrations of Cl^- leads to increase the rate of corrosion of Al-Zn alloy in the basic media.

Chloride ion is bonded chemically in the interface as an initial step of the formation of different mixed oxohydroxo- and chloro-complexes according to the following formula[21]:



Finally the $[\text{AlCl}_6]^{3-}$ complex is produced.

The effect of Cl^- on the polarization curves be more significant in the anodic regions, where can be observe in Figures (5), (6), and (8) anodic polarization observably is marked by two important regions, (A) and (B).

The region (A) show a gradual increase in the current density with applied potential [22]. As soon as region (A) is crossed, a sharp rise in the current density is observed. This is characteristic of region (B).

The break in the curve is due to the onset of pitting as consequence of breakdown of the oxide layer. But in Figures (4), (7) and (9), decrease this phenomena and the increasing in current density is sudden.

5. List of Symbols:

- b Tafel slope (V.decade^{-1})
- E Potential (Volt)
- F Faraday constant (96500 C.mol^{-1})
- i current density (A.cm^{-2})
- R gas constant ($8.314 \text{ J.mol}^{-1} \cdot \text{K}^{-1}$)
- R_p polarization resistance ($\Omega \cdot \text{cm}^{-2}$)
- T temperature (K)

6. Greek letters:

- α transfer coefficient
- η overpotential (Volt)

7. Sub/superscripts:

a anodic
c cathodic
corr corrosion
pass passivity

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تأثير أيونات الكلوريد على سلوك التآكل لسبيكة المنيوم-زنك في محلول هيدروكسيد الصوديوم عند أربع درجات حرارية

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الخلاصة

يهدف البحث الى تصميم ومحاكاة GaussianFSK مرسله باستعمال مركب ترددات من نوع $\Sigma\Delta$ modulator fractional-N وبالمواصفات التالية: مدى الترددات الخارجة (869.9 – 900.4) ميكا هرتز، نسبة تدفق المعلومات (150kb/s)، مدى الفصل بين القنوات (500kHz)، زمن تحويل 1 مايكرو ثانية ومستوى ضوضاء طوري (-85) ديسيبل عند 10000 هرتز من التردد الخارج. تقنيات الدوائر الحديثة ساهمت بشكل فاعل في زيادة التكامل للمرسلات والمستلمات لاشارات الراديو. خواص مثل قلة استبدال القدرة، صغر الحجم وقلة الكلفة هي التي تعتبر المقياس الخاص بالتصميم والتي من خلالها يتم الحكم على اداء النظام. ان التضمين المباشر بواسطة مركب الترددات من نوع $\Sigma\Delta$ (Fractional - N) تم اقتراحه في هذا البحث وذلك لان هذه الطريقة تؤمن الخواص التي نحتاجها مثل قلة استبدال القدرة وصغر الحجم وقلة الكلفة. عملية التضمين من نوع $\Sigma\Delta$ وضعت في دورة قفل الطور الرقمي للسيطرة على القيمة الكسرية لمقسم التردد وبواسطته يتم ازالة الطفيليات مع مستوى طفيليات اقل ومستوى ضوضاء طوري جيد. نوع التضمين (Gaussian FSK) الذي تم استخدامه للحصول على كفاءة طيفية عالية في الاشارة المضمنة. ان التطبيقات لهذه المرسله هي قلة كلفة نقل المعلومات اللاسلكي، انظمة الامان، السيطرة عن بعد بواسطة اشارات (RF) والعداد اللاسلكي.