Corrosion and Corrosion Inhibition of α-Brass by Thiourea

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Abstract

The corrosion behavior and corrosion inhibition of α-brass (65.3% Cu, 34.4% Zn and others 0.3%) in 0.6 mol.dm^{-3} NaCl solution have been investigated using potentiostatic polarization technique, the main results obtained were expressed in terms of corrosion (E_c) and corrosion current (i_c). The research was performed in neutral and slightly acidic media [pH=7 and pH=4] over the temperature range (288-318)K. It was found that the rate of corrosion increases with the increase of acidity and the increase of temperature. The rate of corrosion increased with the increase of temperature in conformity with Arrhenius equation. Values of activation energy (E_a^*), pre-exponential factor (A) and entropy of activation (ΔS^*) have been derived for the corrosion process. Also the thermodynamic quantities (ΔG, ΔS and ΔH) have been determined for the process. The inhibition effect of thiourea on the corrosion of α-brass in chloride solution was studied, and it was found that the addition of thiourea to the chloride solution caused a decrease in the values of corrosion current density and changed to some extent the values of kinetic parameters. The values of (E_a) increases in the presence of thiourea, this means that the decrease of the concentration of thiourea.

Key words: α-brass, thiourea, corrosion inhibition, kinetic and thermodynamic parameters.

Introduction

copper and its alloys are of considerable importance as they form the backbone of modern industries due to their excellent electrical and thermal conductivity. [1, 2] brass has been widely used as tubing material for condensers and heat exchangers in various cooling water system. [3]

Brass is susceptible to a corrosion process known as dezincification in chloride-containing solutions leading to structural failure. [4,5], and this tendency of dezincification increases with the increas zinc content of the brass. [6]

When brass undergoes corrosion, a zinc oxide layer is initially formed which passivites the brass surface, when brass is dipped into a media containing chloride ion, an insoluble film of cuprous chloride is adsorbed on the brass surface. The copper ions can pass into the solution by disproportionation reaction or it can dissolve with the formation of complexes with $CuCl_2^-$ [7]. The formation of stable is possible inside the pores of $CuCl_2^-$ layer. As a result the brass surface became enriched with copper while being impoverished by zinc during corrosion. [8]. These changes make brass surface less resistant to corrosion than copper in chloride containing media. Since, brass is not completely resistant to corrosion, especially in oxygen-containing electrolytes, the use of inhibitor to prevent or minimize corrosion is a standard practice, despite a wide-spread use of inhibitors to control corrosion, still there is a need for more comprehensive and systematic study to understand their mode of action. Precise understanding has become important.

Thiocarbamides [9] were well known for its corrosion inhibition efficiency, especially on transition metals mainly due to the presence of sulphur atom, which has a high electron density and can therefore easily bind to the metal surface. In thiocarbamide, the presence of -group will further contribute stronger binding and enhance inhibition [10]. The effectiveness of any corrosion inhibitor is dependent on the type of the metal, properties of corrosive enviroments as well as the state of inhibitor molecule.

The aim of the present work is to study the effect of thiourea as corrosion inhibitor for dissolution of α-brass in 0.6mol. dm^{-3} solution in neutral and slightly acidic media using potentiostatic polarization measurements. The effect of temperature on the dissolution of α-brass in the absence and presence of the inhibitor was also investigated through the calculations of the kinetic and thermodynamic parameters such as activation energy E_a^*, activation entropy and pre-exponential factor A.

Experimental Part

1.Materials

a- Sodium chloride (analar grade) was used for the preparation of the electrolyte solution of 0.6mol.dm^{-3} (3.5% w/w).

b- Commercial α-brass with the following composition as wt% [65.3% Cu, 34.4% Zn], the remainder being trace amounts of Fe, Sn, Pb, Ni and Al.

c- Thioura (sigma-Aldrich 98%) was used as received.

2.Potentiostatic Polarization Studies

The potentiostatic polarization studies were carried out with apiece of α-brass, which was cut in the form of a disk [2cm diameter and 0.2cm thickness] having an exposed surface area of $1cm^2$ to corrosive medium. The working electrode (α-brass) was abraded mechanically and successively with different grades of emery paper [200, 400,800, 1200 and 2000] and washed with double distilled water. Further, the samples were degreased with acetone and thoroughly washed with double distilled water then dried in air and kept in a desicator until use.

An electrochemical cell with a three electrodes assembly was used to study the electrochemical measurements. Brass specimens with exposed area of 1cm^2, a platinum electrode and silver-silver chloride in staturated were used as working, auxiliary and reference electrodes respectively.

The polarization experiments were carried out using the M lab potentiostat / Galvanostat 200 Germany obtained from bank electronic intelligent controls . M lab was connected to personal computer by a RS 232 serial cable and controlled by computer desktop. The M lab software cares for controlling the potentiostat, recording and processing data. It is provided with electrochemical calculations like tafel line evaluation, re-scaling of the potential and integration.

The experiments were performed in the electrolyte solution of 0.6mol. dm^{-3} at two different values of pH in the absence and presence of different concentrations of the inhibitor (thiourea) over the temperature range mentioned before, the operation program involved sending a set of commands from the computer to the potentiostat, and the polarization experiments were carried for brass specimen at a scan rate of 10mV/s. corrosion current density i_{corr} and corrosion potential E_{corr} . Were determind from the polarization curve in addition to other infromations such as tafel. Slopes, weight loss and penetration loss. In order to test the reproducibility of the results, the experiments were performed in triplicate.

Results and Discussion

1- Polarization Behavior

Figs. 1 and 2 show the anodic and cathodic polarization curves of -brass in 0.6mol.dm^{-3} NaCl solution at four temperatures in the range of (288-318) K and two pH values (pH= 7 and pH =4).

Tafel extrapolation method was used to calculate the corrosion parameters from polarization curves.

The resulting data are displayed in Table (1) and these data show that corrosion current density (i_{corr}) increases with the increase of temperature and nearly corrosion potential (E_{corr}) follows similar manner with the increase of temperature. Also it was noticed that all values of i_{corr} at pH=4 are more than those in pH=7 at all temperatures of study which indicate that α-brass has more tendency to corrode in acidic medium, and this result is enhanced by the values of pentration and weight loss. (Table.1).

Anodic and cathodic tafel slopes show variation in their values which can be attributed to the variation of the rate determing step (r.d.s.) of the metal dissolution reaction (Andoic) and the charge transfer process (desorption or electrochemical desorption) (cathodic) [11].

2- Temperature Dependence of The Corrosion Current Density

The rate of α-brass corrosion (r) (which is expressed by i_{corr}) at a given concentration increased considerably with the rise of temperature. The dependence of the corrosion current density (i_{corr}) on temperature followed Arrhenius equation [12].

$$r = i_{corr} = A_{exp} (-E_a / RT) \ldots\ldots\ldots\ldots(1)$$

which can be expressed in logarithmic form:

$$\log i_{corr} = \log A - \frac{E_a}{2.3RT} \ldots\ldots\ldots\ldots(2)$$

Where A and are respectively the pre-exponential factor and the activation energy of corrosion. A typical linear plot relating values of log to the reciprocal of temperature (1/T) is shown in Figs.3 and 4.

The value of could be derived from the slope of the line, and when the linear plot of Figs. 3 and 4 was extrapolated to log value at 1/T=0, the value of A could be obtained.

Table 2 presents the values of and the pre-exponential factor A for α-brass corrosion in the two pH values 7and 4. It was found that there is direct relation between the values of and A i.e. smiltataneous increase or decrease in and log A for particular system which can be ascribed to the compensation effect which describes the kinetics of catalytic and tarnishing reactions on the metal. [13, 14]

Entropy of activation (ΔS^*) was calculated from the value of A using the relationship.

$$A = \frac{kT}{h}\exp(\Delta S^*/R)\ldots\ldots\ldots\ldots\ldots(3)$$

Where k is boltzman constant, h is plank constant, R is the universal gas constant, and T is the temperature of the solution. The negative values of the entropy of activation (ΔS^*) for α-brass corrosion implies a less in the over-all degrees of freedom throughout the formation of the activated complex for the reaction of α-brass constituent with the negative species (Cl^- & OH^-) leading to the formation of corrosion product [15], when the activated complex result only after considerable arrangements of the structure of reactant molecules, making the complex aless probable structure, ΔS^* is negative, and the reaction will be slower. [16].

3- Thermodynamics of Corrosion of α-brass

The change in Gibbs free energy (ΔG) for the corrosion of the metal specimens in a given pH at given temperature can be estimated from the equation[17]:

$$\Delta G = - nFE_{corr}\ldots\ldots\ldots\ldots(4)$$

Where n (considered to be equal 2) is the number of electrons involved in the anodic process. From the values of   at four different temperatures in the range (288-318)K at the two values of pH (7 and 4), the change in the entropy () of corrosion process could be derived using the well-known thermodynamic relation.

$$-d(\Delta G)/ dT = \Delta S\ldots\ldots\ldots\ldots(5)$$

Utilizing the values of   and , it was possible to calculate the values of the change in the enthalpy for the corrosion process from the relation:

$$\Delta G = \Delta H - T\Delta S\ldots\ldots\ldots\ldots(6)$$

The thermodynamic quantities ΔG, ΔS and ΔH for α-brass corrosion in 0.6mol.dm^{-3} NaCl solution and in the two pH values at four different temperatures (288-318)K are given in Table (3). The obtained results indicate negative values of ΔG that means spontaneous reactions occur.

The enthalpy changes (ΔH) for α-brass corrosion have negative values indicating exothermic reaction while the positive values of ΔS for this corrosion confirm that the corrosion process is entropically favorable.

4- Corrosion Inhibition of α-brass by Thiocarbamide

Fig. 5 (A,B,C,D,E,F) shows the typical polarization curves of α-brass in 0.6mol. dm^{-3} NaCl solution containing three different concentrations of thiourea over the temperature (288-318)K.

Table 4 presents the polarization data ($E_{corr.}$ and $i_{corr.}$) and from these data, it can be noticed that the addition of thiourea caused a decrease in corrosion current densities of α-brass, but this inhibition effect of thiaurea decrease as the concentration of thiourea increased in the range (10^{-2}-10^{-4}) mol.dm^{-3} at all temperatures of study. Thiourea has high inhibition efficiency at low concentration and loses its efficiency at high concentrations. [17] the acceleration of corrosion at higher concentration of thiourea is explained in several different papers [18-20], but no satisfactory explanation is given. The inhibition efficiency of thiourea is controlled by the adsorption of molecular species which increases the inhibition, and the protonated species, accelerating the rate of corrosion. The protonation process is controlled by the charge density on the sulphur atom, lengthening and weakening of the C-S bond and the reactivity of the molecule.

Table 5 shows the values of protection efficiencies (p%) which are calculated from equ.(7), $i_{corr(1)}$ and $i_{corr(2)}$ are corrosion current densities in the absence and presence of the inhibitor respectively.

$$P\% = (i_{corr(1)} - i_{corr(2)} / i_{corr(1)} \times 100 \ldots\ldots\ldots\ldots\ldots(7)$$

5- Effect of Temperature on The Inhibition of α-brass

The effect of temperature on the corrosion rate of α-brass in 0.6mol.dm^{-3} NaCl solutions at (two pH values) in the presence of different concentrations of inhibitor was studied in the temperature range (288-318)K.

As the temperature increases, the rate of corrosion increases and hence the inhibition efficiency of the inhibitor decreases. This is due to the desorption which is aided by the increase of the temperature. This behave proves that the adsorption of the inhibitor on α-brass surface occurs through the physical adsorption. The activation energy E_a and A and entropy of activation ΔS^* for corrosion of α-brass in the presence of different concentrations of inhibitor were calculated from Arrhenius type equation.

$$r= A_{exp} (-E_a/RT) \ldots\ldots\ldots\ldots\ldots..(8)$$

Where the values of (r) was taken to be proportional to the corrosion current density ($i_{corr.}$), A is the pre-exponential factor, E_a is the activation energy and R is the gas constant.

The Arrhenius law is presented as a straight line when log i_{corr} values are plotted against 1/T values Fig. 5 (A.B.C.D.E.F) are estimated from the slopes and intercepts of the plots respectively, and these values are collected in Table 6.

The variation of activation energy in the presence of different concentrations of thiourea would be illustrated as follows; [21] lower values were found in the presence of inhibitor than those without inhibitors i.e. this type of inhibitor retard corrosion at high temperatures but inhibition is diminished at ordinary temperatures. The decrease of E_a with an increase of Thiourea concentration suggest that the energy barrier of corrosion reaction decreases with inhibitors concentration. The higher E_a values in the presence of inhibitor supports the proposed physisorption mechanism, (the system with 10^{-4} mol.dm^{-3} thiourea) unchanged or lower values of E_a in inhibited systems indicate chemisorption mechanism[22].

Conclusion

1. The (α–brass) corrodes in sodium chloride solution by selective corrosion reaction which increases with the increase of acidity and temperature.

2. Thiourea acts as inhibitor for α-brass corrosion, and the inhibition efficiency decreases as concentration of the inhibitor increases.

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Table (1): Data of polarization curve for corrosion of α-brass in 0.6 mol.dm^{-3} NaCl solution at three pH values (4 and 7) over the temperature range (288-318)K.

pH	T/K	i_{corr}/μA.cm^{-2}	-E_{corr}/mV	ba/mV. decade^{-1}	-bc/mV. decade^{-1}	weight loss/ g.m^{-2}. day^{-1}	Penetration loss/mm.year^{-1}
4	288	1.26	238.9	36.5	23.2	0.358	0.0146
	298	1.76	240.3	52.4	185.4	0.551	0.0225
	308	2.91	268.2	51.5	133.4	0.763	0.0312
	318	9.69	264.0	56.4	159.1	1.820	0.0744
7	288	1.22	248.8	57.4	119.5	0.348	0.0142
	298	1.63	257.2	53.9	124.5	0.522	0.0213
	308	2.17	258.9	48.7	120.2	0.618	0.0253
	318	5.85	257.6	43.0	143.2	1.520	0.0622

Table (2): Activation energy (Ea), pre-exponential factor (A) and entropy of activation (ΔS*) for α-brass corrosion in 0.6 mol.dm^{-3} NaCl solution.

pH	**Ea/kj. mol^{-1}**	**-ΔS*/J.K^{-1}.mol^{-1}**	**A/ molecule. cm^{-2}.s^{-1}**
4	47.442	81.835	22.019×10^{31}
7	50.935	74.803	73.703×10^{31}

Table (3): The thermodynamic functions for corrosion of α-brass in 0.6 mol.dm^{-3} NaCl solution over the temperature range (288-318)K in three pH values (4 and 7).

pH	**T/K**	**-ΔG/kj.mol^{-1}**	**ΔH/Kj.mol^{-1}**	**ΔS/j.K^{-1}.mol^{-1}**
4	288	46.107	3.169	171.1
	298	46.377	4.610	
	308	51.762	0.936	
	318	50.952	3.457	
7	288	48.018	-32.408	54.2
	298	49.639	-33.487	
	308	49.967	-33.273	
	318	49.716	-32.480	

Table (4): Values of (-$E_{corr.}$, $i_{corr.}$ and inhibitor effecincy percent) with different concentrations of thiourea at temperature rang (288-318)K in pH=4and 7

pH	**T/K**	**Inhibitor conc. mol.dm^{-3}**	**-E_{corr}/mV**	**-$i_{corr.}$ μA.cm^{-2}**	**IE%**
4	288	0	196.5	1.26	0
		1×10^{-2}	413.3	1.53	-
		1×10^{-3}	320.0	0.85	32.5
		1×10^{-4}	283.0	0.78	38.0
	298	0	248.6	1.76	0
		1×10^{-2}	408.4	1.6	9.0
		1×10^{-3}	326.3	0.9	48.8
		1×10^{-4}	285.0	0.80	54.5
	308	0	268.2	2.91	0
		1×10^{-2}	403.4	2.76	5.1
		1×10^{-3}	335.3	1.72	40.8
		1×10^{-4}	314.5	1.38	52.5

	318	0	264.0	9.69	0
		1×10^{-2}	392.0	5.8	40.1
		1×10^{-3}	339.8	3.42	64.4
		1×10^{-4}	272.1	3.1	68.0
7	288	0	248.8	1.22	0
		1×10^{-2}	417.0	1.02	16.3
		1×10^{-3}	325.7	0.589	51.17
		1×10^{-4}	287.4	0.50	59.0
	298	0	257.2	1.63	0
		1×10^{-2}	403.2	1.3	20.24
		1×10^{-3}	327.6	0.675	58.5
		1×10^{-4}	295.6	0.31	80.98
	308	0	258.9	2.17	0
		1×10^{-2}	407.3	2.21	-
		1×10^{-3}	331.9	1.22	43.7
		1×10^{-4}	311.7	1.12	48.3
	318	0	257.6	5.85	0
		1×10^{-2}	404.6	4.8	17.9
		1×10^{-3}	332.2	2.73	53.33
		1×10^{-4}	298.7	1.68	71.28

Table (5): Values of protection efficies calculated from $i_{corr.}$

pH	**Conc. of thiourea mol.dm^{-3}**	**T/K**	**P% form $i_{corr.}$**
4	1×10^{-2}	288	-21.4
		298	29.8
		308	-8.0
		318	16.4
	1×10^{-3}	288	7.9
		298	31.4
		308	35.8
		318	62.1
	1×10^{-4}	288	37.6
		298	58.7
		308	48.5
		318	23.1
7	1×10^{-2}	288	16.3
		298	11.4
		308	-1.8
		318	31.9
	1×10^{-3}	288	34.0
		298	52.9
		308	43.7
		318	67.6
	1×10^{-4}	288	58.4
		298	55.6
		308	48.3
		318	68.5

Table (6): Activation energy (E_a), pre-exponamtial factor (A) and entropy of activation (ΔS^*) for the corrosion of α-brass in the pH values 4 and 7 in 0.6mol.dm^{-3} NaCl solution and different concentrations of thiourea.

pH	Conce. of thiourea mol.dm^{-3}	E_a/ KJ.mol^{-1}	A/ molecule. cm^{-2}.S^{-1}	-ΔS*/ J.k^{-1}. mol^{-1}
4	1×10^{-2}	51.008	81.411×10^{31}	73.977
	1×10^{-3}	52.765	95.699×10^{31}	72.634
	1×10^{-4}	53.679	118.279×10^{31}	70.875
7	1×10^{-2}	51.765	89.062×10^{31}	73.231
	1×10^{-3}	55.331	196.114×10^{31}	66.677
	1×10^{-4}	66.092	8.41×10^{34}	35.467

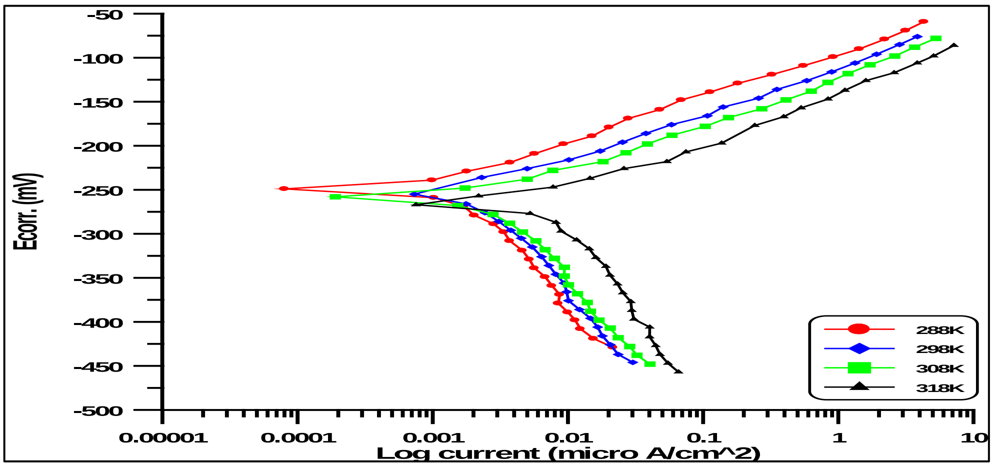

Fig. (1): Polarization curves for the corrosion of α-brass in pH=7 with 0.6mol.dm^{-3} solution at different temperatures in the range of (288-318)K.

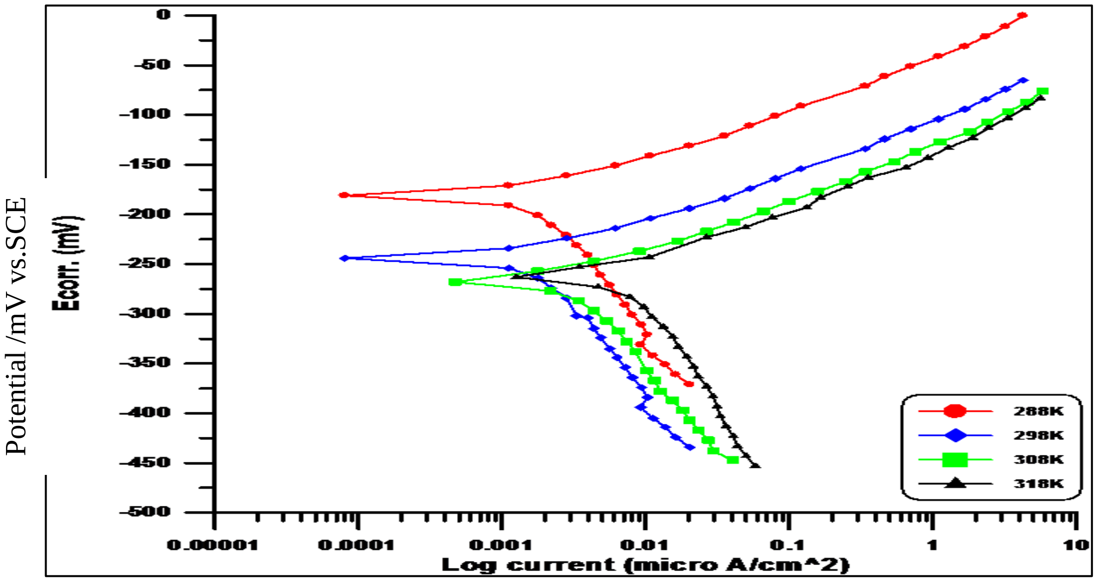

Fig. (2): Polarization curves for the corrosion of α-brass in pH=4 with 0.6mol.dm^{-3} solution at different temperatures in the range of (288-318)K.

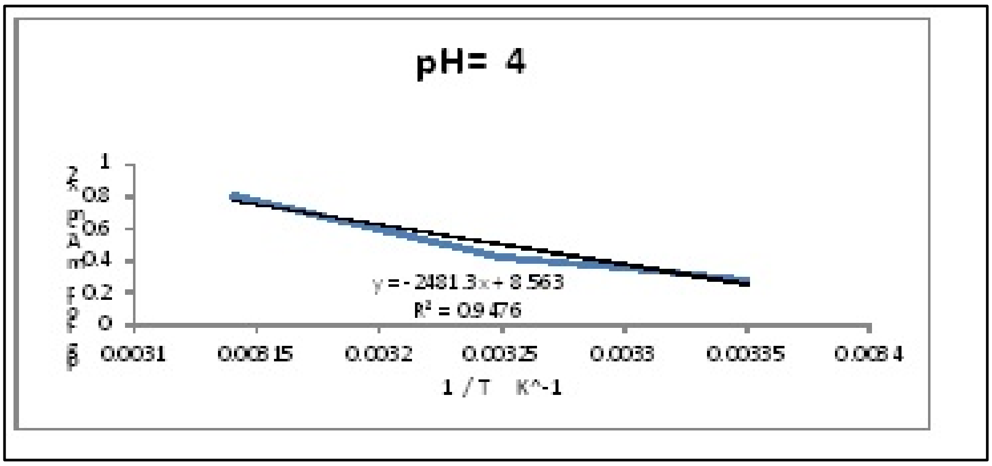

Fig. (3): Arrhenius plots relating log $i_{corr.}$ Vs.1/T for the corrosion of α-brass in 0.6mol.dm^{-3} NaCl in pH=4

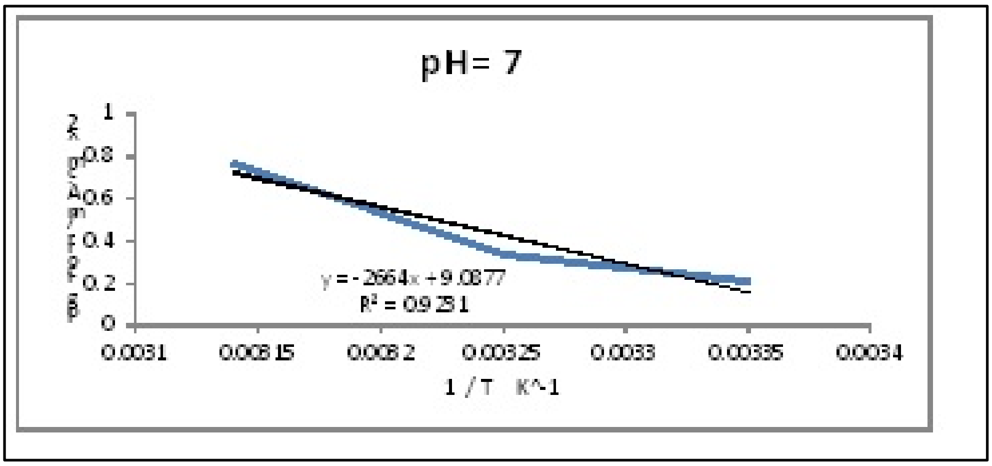

Fig. (4): Arrhenius plots relating log $i_{corr.}$ Vs.1/T for the corrosion of α-brass in 0.6mol.dm^{-3} NaCl in pH=7

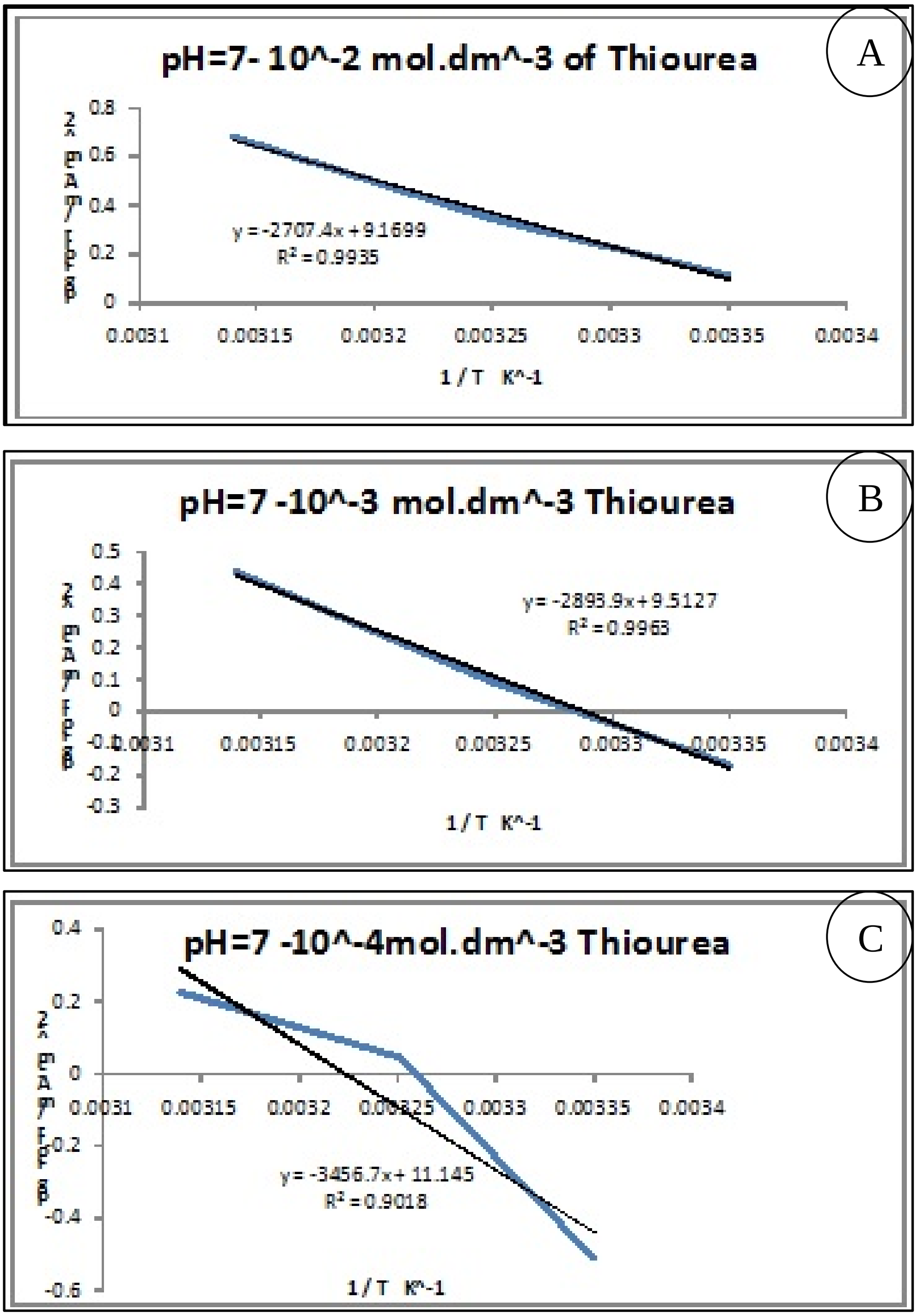

Figure (5 A,B and C): Arrhenius plot, Log i_{corr} as a function of the reciprocal temperature (1/T) for corrosion of α-brass in 0.6 mol.dm^{-3} NaCl solution over the temperature range (288-318)K in pH=7 with three concentration values of thiourea (10^{-2}, 10^{-3} and 10^{-4}) mol.dm^{-3}.

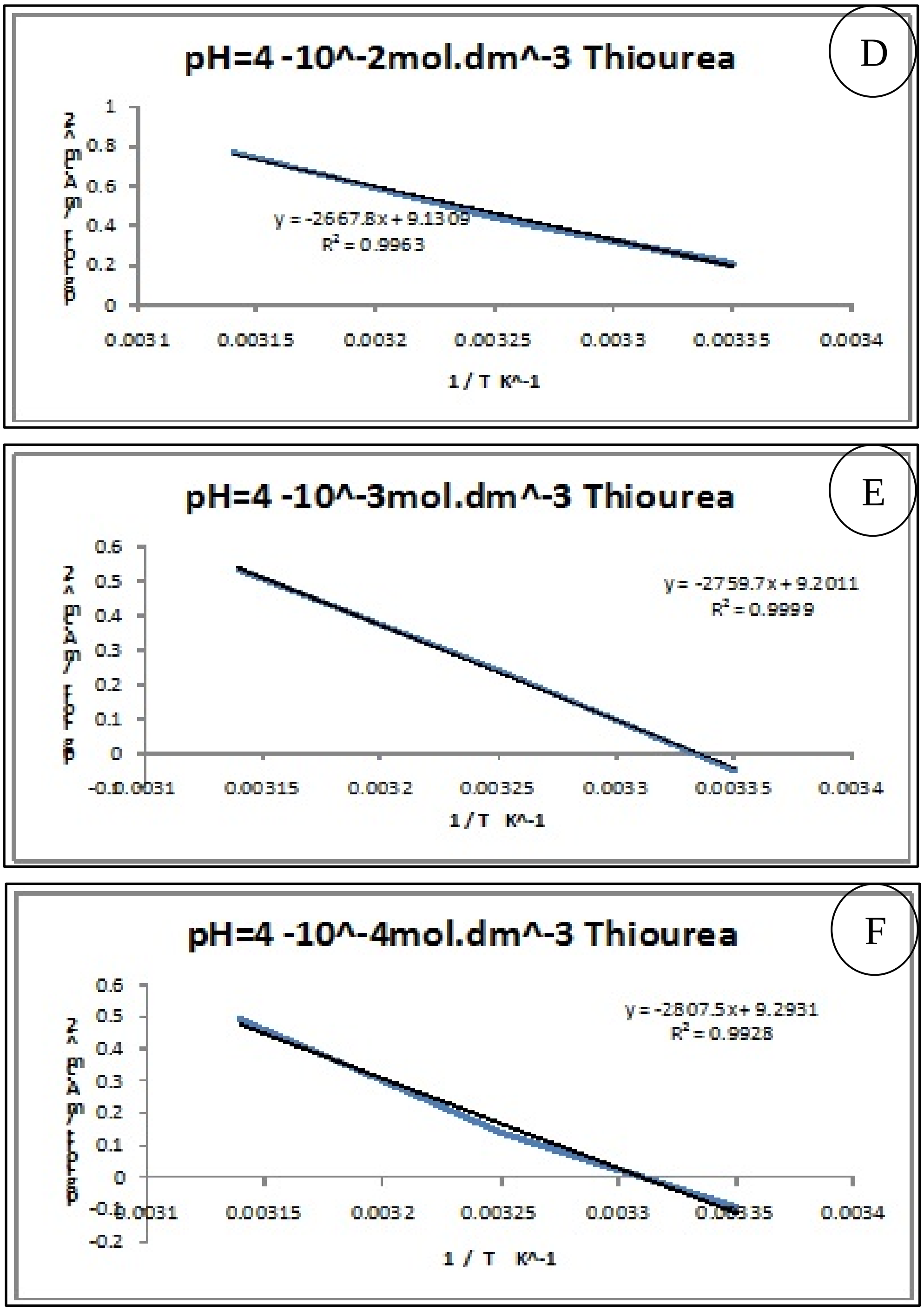

Figure (5 D,E and F): Arrhenius plot, Log i_{corr} as a function of the reciprocal temperature (1/T) for corrosion of α-brass in 0.6 mol.dm^{-3} NaCl solution over the temperature range (288-318)K in pH=4 with three concentration values of thiourea (10^{-2}, 10^{-3} and 10^{-4}) mol.dm^{-3}.

تآكل وتثبيط تآكل سبيكة الفا- براص بواسطة الثايويوريا

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استلم البحث في : 29 آيار 2013 ، قبل البحث في : 4 كانون الأول 2013

الخلاصة

يتناول موضوع البحث دراسة كهروكيميائية لتأكل وتثبيط تاكل سبيكة الفا براص (α-brass) Cu 65.3% ، Zn 34.4% في محلول ملحي لكلوريد الصوديوم بتركيز 0,6 مول. دسم$^{-3}$ باستعمال تقنية المجهاد الساكن، وتم التعبير عن النتائج من خلال تسجيل قيم جهد التآكل (i_c) . درس سلوك التآكل في الوسط المتعادل والقليل الحامضية pH 4 و7 وعلى مدى من درجات الحرارة من 288 الى 318 كلفن. لوحظ ان سرعة التآكل تزداد بزيادة الحامضية وزيادة درجة الحرارة، اذ خضعت حركية تفاعل التآكل لمعادلة ارينيوس التي من خلالها حسبت قيم طاقة التنشيط (E_a^*) ومسبوق المقدار الاسي (A) وانثروبي التنشيط (ΔS^*) كما امكن حساب الكميات الثرموديناميكية لتفاعل التآكل ($\Delta G, \Delta S, \Delta H$) كما اشتمل البحث على دراسة استعمال مادة الثايويوريا مثبطاً لعملية تآكل سبيكة الفا براص في محلول كلوريد الصوديوم بالتركيز المشار اليه اعلاه، واتضح ان اضافة الثايويوريا عملت على تقليل سرعة التآكل من خلال قيم تيار التآكل المتناقصة، ولوحظ زيادة قيم طاقات التنشيط عند اضافة الثايويوريا وهذا يعني ازدياد الحاضر الطاقي لعملية التآكل وكانت هذه الزيادة في علاقة عكسية مع تركيز الثايويوريا.

الكلمات المفتاحية: الفا براص، ثايويوريا، تثبيط تآكل، معلمات حركية وثرموديناميكية.