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Inhibition of Aluminum Alloy 7025 in Acid Solution Using Sulphamethoxazole

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Abstract

Corrosion behavior of aluminum alloy 7025 was investigated in hydrochloric acid (pH=1) containing 0.6 mol.dm^{-3} NaCl in the existence and absence of diverse concentrations of sulphamethoxazole as environmentally friendly corrosion inhibitor over the temperature range (298-313)K. Electrochemical polarization method using potentiostatic technique was employed. The inhibition efficiency has been raised with increased sulphamethoxazole concentration but lessened at temperature increases. The highest efficiency value was 96.5 at 298 K and 2 x10^{-4} mol.dm^{-3} concentration of sulphamethoxazole. The sulphamethoxazole adsorption was agreed with Langmuir adsorption isotherm. Some thermodynamic parameter ($\triangle G_{ads}$) and activation energy (E_a) were determined to demonstrate the process of corrosion inhibiting. The kinetic parameters were calculated using Arrhenius equation. An appropriate mechanism has been suggested for aluminum corrosion in acidic medium. The results show that sulphamethoxazole is a mixed type inhibitor. The surface morphology of uninhibited and inhibited samples was investigated using an optical microscope.

Keywords: Corrosion inhibition, Aluminum, Sulphamethoxazole, Free energy, Adsorption mechanism.

Introduction

Corrosion is a physically natural activity which causes degradation of the material's features throughout its electrochemical or chemical interacting with the adjacent atmosphere. The related losses of corrosion are either direct or indirect losses. Direct losses are as a result of the substitution of rusted parts, accomplishment, and protection of corroded structures, while indirect losses are owing to the blackout of industrial plants, efficiency, and product failures.

Reported studies about corrosion have as well been essential as a result of the cumulative requirement attentiveness to protect the realm's metal resources [1]. Currently, more consideration was to regulate the metallic corrosion, because of growing consumption of metals in entire technology fields.

Corrosion investigations of aluminum and aluminum alloys have received substantial consideration by scholars as a consequence of their widespread manufacturing and commercial issues [2]. Aluminum and aluminum alloys have arisen as alternative constituents in numerous uses and in several chemical processing productions. By reason of their extensive applications, they recurrently come with acids or bases throughout de-scaling, pickling, electrochemical etching and expansively employed in chemical process productions. The majority of testified researches were accomplished on corrosion of different metal as well as alloys in HCl and H$_2$SO$_4$ mediums [3-5].

Hydrochloric acid typically employed for chemical and electrochemical etching or pickling of aluminum objectives as a part of investigations with aluminum corrosion occurrence in hydrochloric acid medium and corrosion regulating utilizing green inhibitor.

As corrosion inhibitors, some organic or inorganic

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substances have been used to increasing resistance of metals and alloys against corrosive environmental factors and decreasing the corrosion rate [6-9]. The toxicity of organic and inorganic inhibitors unacceptable for the environment and human health. Thus it is necessary to find cheap and environmentally safe corrosion inhibitors [10,11]. Studies are directed towards the exploration for eco-friendly corrosion inhibitors which are ecologically acceptable. Recently drugs are used as corrosion inhibitors [12-14]. The drug uses as corrosion inhibitors for metal corrosion are the best choice because of their eco-environmental nature. Drugs are nontoxic and cheap, so most of them can be produced from natural materials thus they would be insignificant negative influences on the environment. Accordingly, it would be a good alternative for the previous traditional toxic inhibitors [15, 16]. The drugs selection as corrosion inhibitors depends on (O, N and S) as effective centers and they can be produced and purified straightforwardly [17-20].

This study aims to characterize the influence of sulphamethoxazole as corrosion inhibitors for aluminum alloy 7075 in acidic medium (pH=1) containing (3.5% w/w) sodium chloride at a temperatures range (298-313) K.

Experimental

Materials

The specimens used in this investigation were taken from aluminum alloy 7025 sample of the following composition given in Table- 1.

Table 1

The chemical composition of aluminum alloy 7025.

Zn	Fe	Cu	Mn	Mg	Cr	Ti	Si	Al
5	0.4	0.2	0.5	1.6	0.3	0.1	0.3	Bal.

Solutions have been formulated from analytical grade chemicals and particularly disinfected water. Hydrochloric acid has gotten from Fluka Company, whereas sulphamethoxazole has gotten from Aldrich chemical Co. Ltd. All measured parameters have achieved in acidic solution (pH=1) having 3.5% NaCl that possesses an equivalent level to that of saltwater.

Methods
Potentiostatic polarization measurements

The measured potentiostatic polarization results have been implemented in a conservative three-electrode glass cell with functioning volume 1000 ml

utilizing Wenking M lab potentiostat with a platinum counter electrode and silver-silver chloride as a reference electrode.

An employed electrode was a sheet with a circular shape (2 cm diameter and 2 mm thickness) which previously were grinded with emery papers of different graded (200, 400, 800, 1200 and 2000). Then they were polished mechanically with diamond paste using a smooth cloth to a surface mirror. The exposed surface area to the aggressive medium was 1cm^2.

Silver-silver chloride as a reference electrode and Pt counter electrode has been employed. Measured polarization records have been implemented at a potential from -200 mV to +200 mV at a scan rate of 2mV/s after steady state potential had been established in cathodic or anodic direction. The entire measurements have been achieved in an acidic solution (pH=1) of 3.5% NaCl in the lack and existence of three distinct concentrations of the inhibitors.

The investigational results have been reproducible, and each experiment has been carried out at least twice.

Results and Discussion

Polarization curves

Figure- 1 shows the anodic and cathodic polarization curves of aluminum alloy 7025 in acidic solution (pH=1) of 0.6 mol dm^{-3} NaCl (3.5% W:W) at four temperatures in the range (298-313)K.
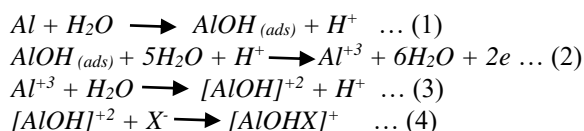
Tafel extrapolation method was used to calculate the corrosion parameters (E_{corr}), (i_{corr}), (b_c) and (b_a) from the polarization curves.

The resetting data were displayed in Table- 2. These results show that corrosion current density (i_{corr}) increases with cumulative temperature. The corrosion potential (E_{corr}) is nearly more negative with increasing temperature the negative shifting in the corrosion potential (E_{corr}) with the increased temperature specifies that the anodic method is highly influenced as compared with the cathodic process. This performance is consistent with other published results [1,2] which proposed the dependence of (i_{corr}) and (E_{corr}) on solution parameters. The change in (b_c) and (b_a) reflects the mechanism of corrosion process.

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The electrochemical cathodic and anodic reactions of aluminum in acidic solution can be described as follows [21]:

$$Al + H_2O \longrightarrow AlOH_{(ads)} + H^+ \quad \dots (1)$$
$$AlOH_{(ads)} + 5H_2O + H^+ \longrightarrow Al^{+3} + 6H_2O + 2e \dots (2)$$
$$Al^{+3} + H_2O \longrightarrow [AlOH]^{+2} + H^+ \quad \dots (3)$$
$$[AlOH]^{+2} + X^- \longrightarrow [AlOHX]^+ \quad \dots (4)$$

Consequently, formed soluble complex ion causes the meta dissolution. Cathodic and anodic slops for 7025 aluminum alloy under (0.6 mol.dm^{-3}) and pH of 1 conditions without and with three different concentrations of inhibitor at a temperature range (298-313) K were studied as in Figures 1 and 2 respectively. In the presence of sulphamethoxazole, the corrosion current densities reduced as a concentration of inhibitor augmented. Table- 3 shows the values inhibition efficiencies (*IE %*) which could be calculated from equation [22]:

$$IE\% = [(i^\circ_{corr} - i_{corr}) / i^\circ_{corr}] \times 100 \dots (5)$$

where i°_{corr} and i_{corr} are corrosion current densities without and with the inhibitor respectively. A maximum efficiency value was found to be 96.5 at 298 K and 2×10^{-4} mol.dm^{-3} of sulphamethoxazole.
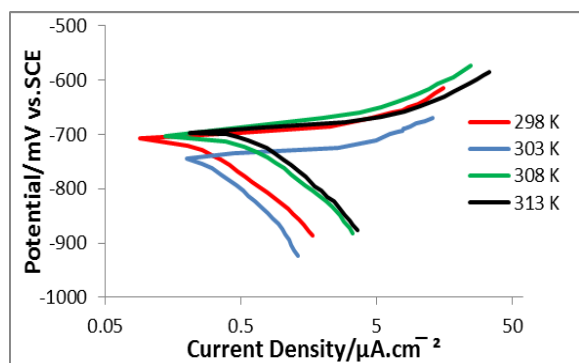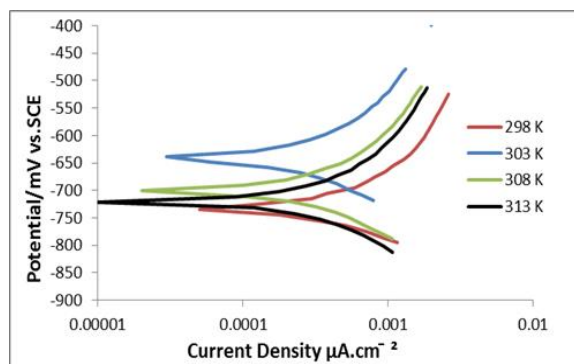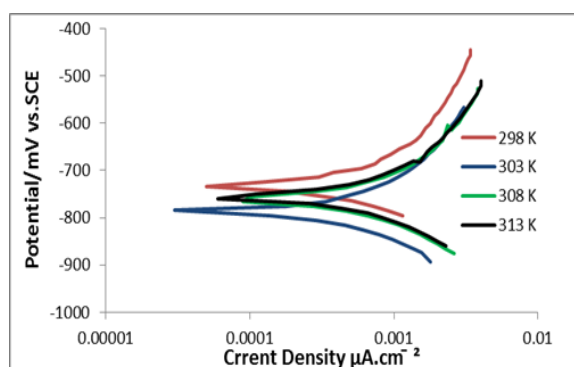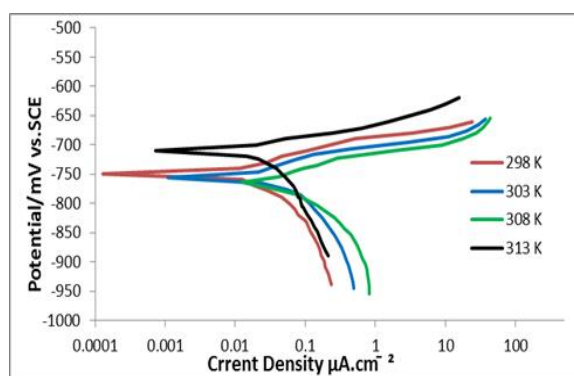

Fig. 1. Polarization curves of aluminum alloy 7025 corrosion in 0.6 mol.dm^{-3} NaCl solution at pH=1 and temperatures range (298-313) K.

a

b

c

Fig. 2. (a, b and c) Polarization curves of aluminum alloy 7025 corrosion in 0.6 mol.dm^{-3} NaCl solution at pH=1 and temperatures range (298-313) K with concentrations of sulphamethoxazole (0.4×10^{-4}, 1×10^{-4} and 2×10^{-4}) mol.dm^{-3}.

Table 2

Data of polarization curve of aluminum alloy 7025 corrosion in 0.6 mol.dm^{-3} NaCl solution at pH=1 and temperatures range (298-313) K.

T (K)	i_{corr} (μA.cm^{-2})	$-E_{corr}$ (mv)	b_c (mv.decade^{-2})	b_a (mv.decade^{-2})	Weight loss (g.m^{-2}.day^{-2})	Penetration loss (mm.year^{-1})
298	710.8	223.14	175.1	27.0	1.80 x 10^{-1}	2.43
303	745.2	278.06	218.7	27.4	2.24 x 10^{-1}	3.02
308	701.6	367.96	128.7	29.8	2.96 x 10^{-1}	4.00
313	698.0	397.36	169.0	28.4	3.20 x 10^{-1}	4.32

Table 3

Values of E_{corr}, i_{corr}, inhibitors efficiency and concentrations of sulphamethoxazole of aluminum alloy 7025 corrosion in 0.6 mol.dm^{-3} NaCl solution at pH=1 and temperatures range (298-313) K.

T (K)	Inhibitor conc. (mol.dm^{-3})	E_{corr} (mv)	i_{corr} (mA.cm^{-2})	Θ	IE% from i_{corr}
	0	710.8	223.14	-	0
298	0.4 x10^{-4}	734.6	59.90	0.730	73.0
	1.0 x10^{-4}	721.0	30.43	0.860	86.0
	2.0 x10^{-4}	746.6	7.74	0.965	96.5
	0	745.2	278.06	-	0
303	0.4 x10^{-4}	639.8	84.59	0.695	69.5
	1.0 x10^{-4}	785.1	42.77	0.846	84.6
	2.0 x10^{-4}	756.7	38.13	0.860	86.0
	0	701.6	367.96	-	0
308	0.4 x10^{-4}	702.3	139.64	0.620	62.0
	1.0 x10^{-4}	762.7	96.44	0.737	73.7
	2.0 x10^{-4}	760	60.38	0.835	83.5
	0	698	397.36	-	0
313	0.4 x10^{-4}	722	177.64	0.55	55.0
	1.0 x10^{-4}	758.3	170.37	0.57	57.0
	2.0 x10^{-4}	720	95.27	0.76	76.0

Effect of temperature

Arrhenius suggested the famous equation that correlates the temperature variation with a rate of corrosion. The influence of increasing temperature on the corrosion rate of Al alloy 7025 which is expressed by (i_{corr}) at a given concentration as the following formula [23]:

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$$ln\ (i_{corr}) = ln\ A - (Ea^*/RT)\ \dots(6)$$

where A is the frequency factor, Ea^* represents the activation energy, R is the gas constant and T is the absolute temperature. Equation (5) predicts that plotting of $ln(i_{corr})$ versus $1/T$ should be linear as we tentatively detected as in Figure- 3. The line slope gives as $(-Ea^*/RT)$, whereas the intercept of the line extrapolated to ($1/T$=0) gives lnA. The value of activation energy equal to (57) kJ mol^{-1}, Table- 4.

Entropy of activation was calculated from the value of A using the relationship [24]:

$$A = (kT^*/h)\ exp\ \triangle S^*/R\ \dots (7)$$

where k is Boltzmann constant, h is Plank constant and T is the temperature of the solution. The value of the entropy of activation was found to be (-27.7) JK^{-1}.mol^{-1}. The calculated values Ea^* and $\triangle S^*$ are depicted in Table (4). In the sulphamethoxazole existence, intensification in Ea^* values has been noticed.

This increase in Ea^* values in the inhibitor presence can be illustrated as follows [25]. Higher magnitudes of Ea^* were found in the presence of inhibitor as compared with those without inhibitor. For instance, the addition of inhibitor raises the energy impediment for the corrosion activity emphasizing the electrostatic character of the inhibitors adsorption on Al alloy 7025 surface.

The values of activation entropy $\triangle S^*$ are negative with and without inhibitor indicating the influential rate step for the stimulated complex for the association instead of the dissociation step. In the inhibitor presence, $\triangle S^*$ moves toward negative values as in Table- 4 that signifies the adsorption process is along with a reduction in entropy. Namely, it means that there an increase in the order going from reactants to actuated complex [26].
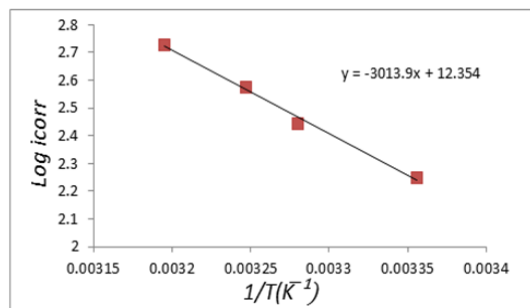

Fig.3. Arrhenius plot of aluminum alloy 7025 corrosion in 0.6 mol.dm^{-3} NaCl solution at pH=1 and temperatures range (298-313) K.

Table 4

Values of activation energy (Ea^*), pre-exponential (A) and entropy of activation ($\triangle S^*$) of aluminum alloy 7025 corrosion in 0.6 mol.dm^{-3} NaCl solution at pH=1 without and with different concentrations of sulphamethoxazole.

Inhibitor conc. (mol.dm^{-3})	Ea^* (kJ.mol^{-1})	$-\triangle S^*$ (J.K.mol^{-1})	A (molecule cm^{-2}.S^{-1})
-	57.72	12.27	1.361 x10^{36}
0.4 x10^{-4}	60.51	11.64	1.437 ×10^{36}
1 x10^{-4}	87.09	71	3.280 ×10^{40}
2 x10^{-4}	116.4	160.4	1.685 ×10^{45}

Adsorption isotherms

Adsorption isotherms make available elementary facts about the interactive nature between the inhibitor and Al alloy 7025 through applying some adsorption isotherm models. Dual foremost kinds of interaction can depict the inhibitors adsorption on the metal surface that are physisorption and chemisorption, depending on the chemical inhibitor structure, the nature of the corrosive medium, and the charge and corrosive metal type.

In aqueous solutions, the metal surface is invariably enclosed with adsorbed water molecules. For that reason, the inhibitor molecules adsorption from an aqueous solution is a quasi-subsisted method, and the inhibitors that can be adsorbed powerfully on the metal surface will delay the dissolution reacting of the immersed metal into the corrosive media. The corrosion mechanism inhibition is feasibly clarified by adsorption behavior based on the calculated coverage degree [25].

The surface coverage as in Table- 3 and the inhibitor concentrations have investigated as a result of fitting to numerous isotherms like Langmuir, Tempkin as well as Freundlich. Nevertheless, the

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finest fit has gotten with Langmuir isotherm as presented in Figure- 4 which is specified by [22]:

$$C_{inh}/\Theta = 1/k_{ads} + C_{inh} \dots (8)$$

where k_{ads} stands for the equilibrium constant of the adsorption/desorption processes, and it reveals the inhibitor molecules affinity towards surface adsorption positions.

From the straight line intercepts on the C_{inh}/Θ axis as in Figure-4, it causes the equilibrium constant of the adsorption / desorption of sulphamethoxazole processing as depicted by Table- 5. The significant magnitude of k_{ads} shows that the acid molecule has resilient adsorption onto Al alloy 7025 surface. Nevertheless, k_{ads} lessened with the increased temperature signifying that adsorption of sulphamethazol onto alloy surface was favorable at lower temperatures.
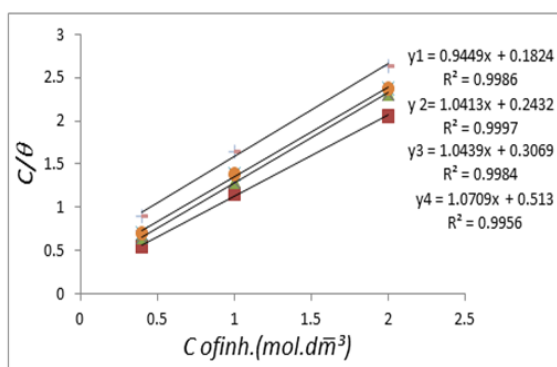

Fig.4. Langmuir adsorption plots of sulphamethoxazole onto aluminum alloy 7025 in 0.6 mol.dm^{-3} NaCl solution at pH=1 and temperatures range (298-313) K.

For the adsorption/desorption, the equilibrium constant k_{ads} was in accordance with the standard free energy of adsorption by [26]:

$$\Delta G° = -RT \, ln(55.5 \, k_{ads}) \dots (9)$$

where R is the universal gas constant, T is the absolute temperature, and 55.5 represent the water concentration in solution.

The standard free energy of adsorption has been evaluated and specified in Table- 5. In general, the values of $\Delta G° \approx$ 26-28 kJ mol^{-1} or less negative values are related with an electrostatic interacting amid charged metal surfaces or more negative values have charge distribution or transfer from inhibitor molecules to the metal surface to generate a coordinate covalent bond. Consequently, it is determined that adsorption of sulphamethoxazole onto Al 7025 alloy surface takes place throughout chemical as well as physical adsorptions.

The 1st step in the adsorption of sulphamethoxazole on the alloy surface typically contains the adsorbed replacement of single or more water molecules on the alloy surface.

$$inh_{sol} + H_2O_{ads} = inh_{ads} + H_2O_{sol} \dots (10)$$
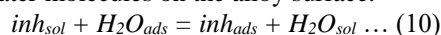
The inhibitor sulphamethazol may subsequently unite with newly produced metal ions (Al^{+3} or other metal elements in the alloy) on the 7025 alloy surface forming metal inhibitor complex [27]:

$$Al \rightarrow Al^{+3} + 3e \dots(11)$$
$$Al^{+3} + inh_{ads} \rightarrow [Al\text{-}inh]_{ads} \dots(12)$$
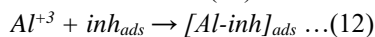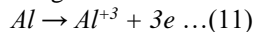
Practical details about corrosion inhibition mechanism are given by the magnitudes of thermodynamic parameters for the adsorption of the inhibitor.

$\Delta G°_{ads}$ has been thermodynamically by the standard enthalpy $\Delta H°_{ads}$ and standard entropy $\Delta S°_{ads}$ based on [28] :

$$\Delta G°_{ads} = \Delta H°_{ads} - T\Delta S°_{ads} \dots(13)$$

The adsorption standard enthalpy is possibly computed based on Van't Hoff formula:

$$lnk_{ads} = - (\Delta H°_{ads} / RT) + Constant \dots(14)$$

A plot of lnk_{ads} versus $1/T$ provides a straight line as presented in Figure-5. The slope of the straight line is equal to $- (\Delta H°_{ads} / RT)$. The negative magnitudes of $\Delta H°_{ads}$ show that the adsorption of inhibitor molecules is an exothermic or endothermic process for sulphamethoxazole. In general, an exothermic adsorption process implies either chemisorption or physisorption whereas endothermic process attributes to chemisorption. The $\Delta G°_{ads}$ values in the inhibitor existence are negative that is gone with exothermic adsorption process. $\Delta S°_{ads}$ of the inhibition process can be calculated from equation (15), according to:

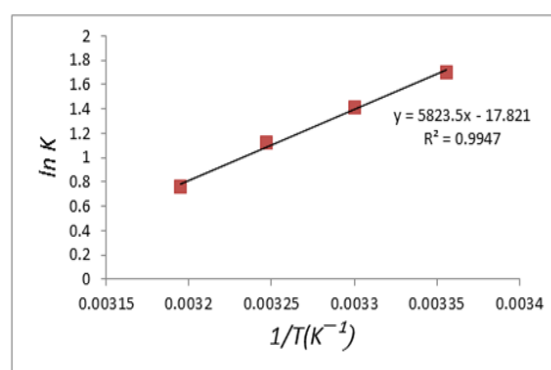$$\Delta S°_{ads} = (\Delta H°_{ads} - \Delta G°_{ads}) / T \dots(15)$$

Fig. 5. Van't Hoff plot of aluminum alloy 7025 in 0.6 mol.dm^{-3} NaCl in a presence of sulphamethoxazole at pH=1 and temperatures range (298-313) K.

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Table 5

Values of equilibrium constant adsorption/desorption, standard free energy, enthalpy and entropy of adsorption onto aluminum alloy 7025 in 0.6 mol.dm^{-3} NaCl solution at pH=1 in a presence of sulphamethoxazole.

| T (K)	k_{ads} (mol^{-1})	$-\Delta G_{ads}°$ (kJ.K.mol^{-1})	$\Delta H_{ads}°$ (kJ.K.mol^{-1})	$\Delta S_{ads}°$ (kJ.K.mol^{-1}.K^{-1})
298	5.4824	14.166		0.1149
203	4.1118	13.679		0.1146
			-48.416	
308	3.2583	13.309		0.1139
313	1.9493	12.188		0.1157

Optical microscopy measurements

To examine the influence of inhibition on the surface morphology of aluminum alloy 7025 surface examination was carried out using optical microscope type of (Nikon Eclipse ME 600, Japan) with and without immersion in the aggressive solution in the lack and existence of the finest concentration of sulphamethoxazole at 25°C. The specimens have been cleaned gradually with water. Afterward, they have dried thoroughly and examined without further actions. Fig. 6 a has depicted the images of aluminum alloy 7025 surface that immersed in the aggressive solution with and without adding 1x10^{-2} mol. dm^{-3} of sulphamethoxazole. It can be observed that the specimen surface has been intensely smashed in lack of inhibitor as a result of metal dissolution in the aggressive solution as depicted in Fig. 6 b.
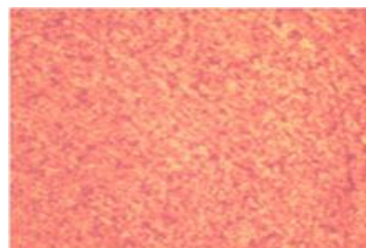
Nevertheless, no cracks and pits are detected in the micrograph after adding inhibitors to the aggressive solution as in Fig. 6 c. Adsorbed inhibitor molecules on effective positions of Al with a smoother surface has detected as compared with the treated surface with uninhibited acidic chloride solution.
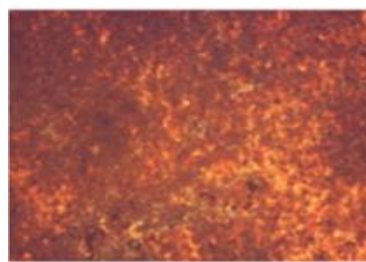
b

c

Fig. 6. Optical microscope micrographs of a: Polished surface of aluminum alloy 7025, b: Surface of aluminum alloy 7025 after the immersion in the corrosive medium and c: Surface of aluminum alloy 7025 after the immersion in the corrosive medium a presence of sulphamethoxyzole.

Conclusions

The results which obtained from this study was confirmed that sulphamethoxazole is good inhibitor for corrosion of aluminum alloy 7025 in acid medium in presence of 0.6 mol.dm^{-3} NaCl . Tafel plots of the inhibitor suggested that sulphamethoxazole acts as mixed type inhibitor. So, the inhibition process is due to physical adsorption of sulphamethoxazole molecules on Al alloy 7025 surface. The protection efficiency increased with the increase of inhibitor concentration. The highest value reaches 96.5% at 2×10^{-4} mol.dm^{-3} of sulphamethoxsazole.

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