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Corrosion inhibitor of carbon steel in 3.5%NaCl solution with Schiff base compounds

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Abstract--In this work, synthesized N₄,N₄'-bis(2, 3, 4 nitro benzylidene) biphenyl-4-4'-diamine(B₁-B₃) , was tested as an inhibitors in controlling the corrosion of carbon steel in NaCl 3.5% solution by using open circuit potential (OCP), at four different temperatures (293, 303, 313 and 323 K). Furthermore, the surface morphology was investigated using the Atomic force microscopy (AFM). The effect of using different Schiff bases and temperature was also investigated. Schiff bases was synthesized and characterized via using. Fourier Transform Infrared Spectroscopy (FT-IR) and Atomic Force Microscope (AFM) characterized . The experimental results shown that Schiff bases can consider as an excellent corrosion inhibitors for carbon steel in NaCl 3.5% solution, by presence of Schiff bases the corrosion of carbon steel was significantly suppressed. The inhibition efficiency is decreased with the increases of temperature. Kinetic and thermodynamic activation parameters (E_a, A, ΔH*, ΔS* and ΔG*) were calculated.

Keywords--Polarization, inhibition, Carbon steel, Schiff base, Corrosion potential.

1. Introduction

The purpose of using of chemical inhibitors to decrease the rate of corrosion process. Inhibitors are chemicals which reacted with metallic surface, or with the environment, this surface is exposed for giving the surface a certain level of protection. Inhibitors are worked by adsorbing themselves on the metal surface, protected the metal surface by forming a film. Some of these are included in a protective coating formulation. Inhibitors can be decreased the corrosion rate by [1]:

* Increasing anodic or cathodic behavior (Tafel slopes). * Increasing electrical resistance of the alloy or metal surface. * Decreasing the diffusion or movement of the ions into the metal surface. The anodic protection involves passivation of the protected metal. The passive film formed on the metal surface by applying of electrical current. The passive film protect the metal from dissolution and this film is insoluble in the environment that is formed. The passivation makes metal non- reactive and resistant to corrosion [2]. The anodic protection is used when the cathodic protection and coatings methods are not provided protection against corrosion [3]. The main principle of cathodic protection is to impress an external current on the material, which forces the electrode potential down to the immune region, or, for protection against localized corrosion, below a protection potential. In other words, the material made the cathode in an electrochemical cell is polarized cathodically as illustrated by potentiallog current curves [4]. schiff bases Compounds that contain a carbon and nitrogen atom connected with double bond with either an aryl or alkyl group attached to the nitrogen atom forming imine groups. The first one who reported them was Hugo Schiff in 1864, it is also called azomethines and its general formula is shown in Figure (1).

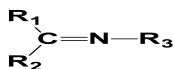
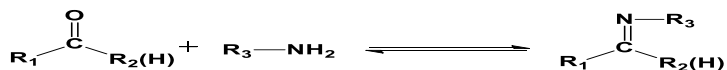


Figure (1): Structure of Schiff bases

Were R is a proton or an aliphatic or aromatic organic compound [5].

Schiff bases are formed by reaction of primary amines with an aldehyde or ketone in suitable conditions forming an imine group which exploits a large range of organic compounds like oxazolidinone, thiazolidinone, tetrazole, amidazolinone and others by ring closer [6], Equation (2).

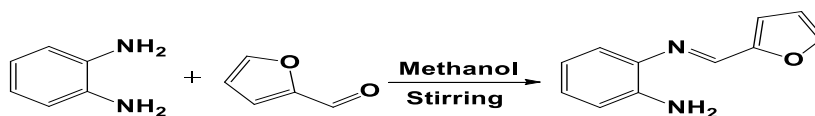


Equation (2): Preparation of Schiff bases

Physically, it can be present in crystalline or oily forms but they are similar in solubility as they dissolve in organic solvents and insoluble in water. Its weak base enables it to form salts with acids in an anhydrous medium, most of these compounds are stable in alkaline solution [5].

Studies are performed on Schiff base metal complex because of their various chemical and physical properties, in addition to using Schiff base in color photography [7]. It is also show analgesics, inflammatory and antimicrobial activities [8].

Omyma A.M.Ali *et al.* (2015) [9] prepared (E)-N1-(furan-2-ylmethylene)benzene-1,2-diamine by stirring furfural with O-phenylene diamine (1:1) at room temperature in the presence of methanol as a solvent, Equation (3).



Equation (3): Preparation of furfural Schiff bases

2. Experimental part

Preparation of specimen

The composition used of carbon steel specimens were (0.36-0.42)% C, (0.15-0.30)% Si, (1.00-1.40)% Mn, 0.05% P, 0.05% S, 0.50% Cu, 0.20% Ni, 0.20% Cr and (96.88-97.49)% Fe with size 1cm diameter specimen were used for method of polarization. The samples were polished with belt grinding polishing machine successively and also with different grit emery papers (80, 150, 220, 240, 320, 400, 1000, 1200 and 2000) in sequence, in the end the specimens were washed with distilled water and dried[10,11].The test samples were prepared by dissolving 5mg of from the prepared derivatives Schiff bases in 2 ml of carbonate. The prepared solution was mixed with sodium silicate then carbon steel coated with preparation fixed then the plate was exposed to medium of Nacl 3.5% solution to measure the corrosion efficiency of these inhibitors [12]. The potentiostat system as shown in Figure (2) that consists of the main computer, the potentiostat that controls the cells of the three electrodes, the thermostat that controls the temperature of the solution and the voltage stabilization device (galvanostate), a magnetic stirrer, the corrosion cell consists of two containers, an external container, and an inner container made of Pyrex glass and has a size of 1L. The electrochemical cell for corrosion consists of three electrodes, the first is the working electrode which consists of carbon steel with an area of 1cm² and the second is the auxiliary electrode that is made of platinum of length 10 cm and the third is the reference electrode that consists of silver chloride (Ag/AgCl,3.0M KCl). The working electrode was immersed in the sample solution for a period of half an hour to stabilize the open circuit, Tafel polarization curves were obtained automatically from -200 mV versus open circuit potential (OCP) to +200mV versus OCP with scan rate of 2.0 mvs⁻¹, all experiments were conducted at (293,303,313,323) K, inhibitor Tafel lines of potential versus I_{corr} in a logarithm scale were plotted and corrosion current density (I_{corr}) and corrosion potential (E_{corr}) were determined in the absence and presence of inhibitor [13].

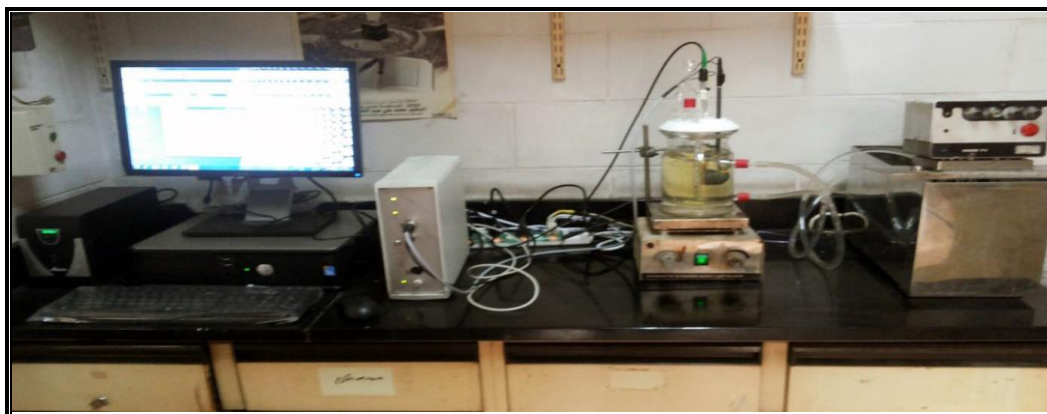
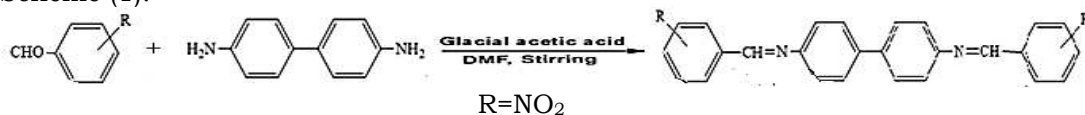


Figure (2): The complete potentiostate system setup.

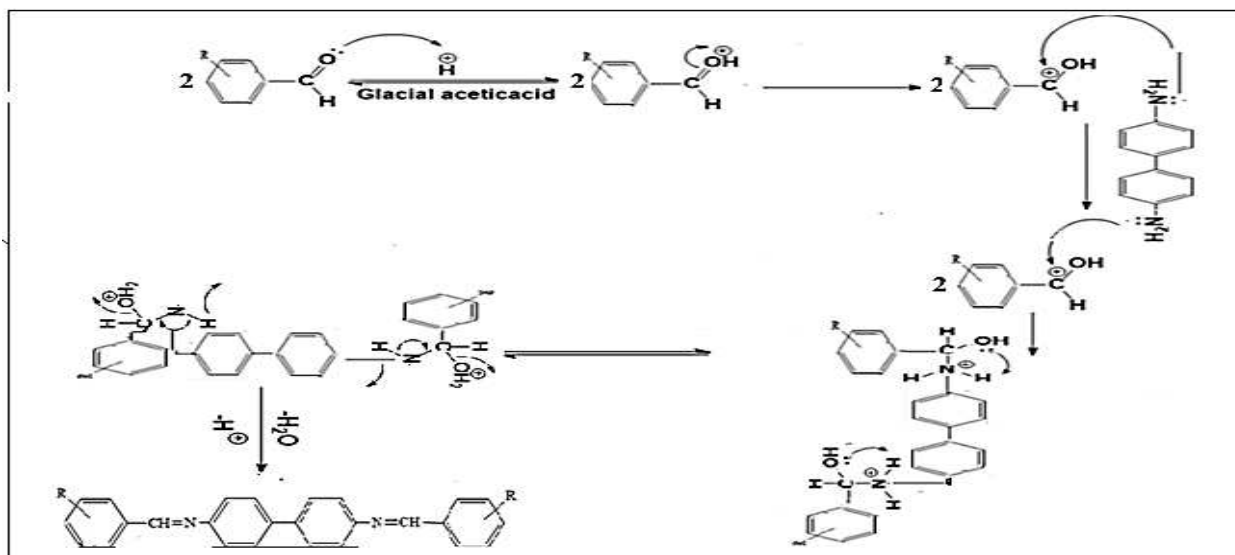
3. Results and Discussion

3. 1 Mechanism for preparation of compounds (B₁-B₃)

These compounds are made by synthesis a nucleophilic addition of several substituted amines to aldehyde in the presence of glacialacetic acid as shown in Equation (4). The formation first step is carbocation which is attacked by the nitrogen lone pair in the amine forming the medium hemi aminal (β -hydroxy amine) which is stable by losing a water equal to form Schiff bases [14] as shown in Scheme (1).



Equation (4): Preparation of compounds (B₁-B₃).



Scheme (1): Preparation mechanism of compounds (B₁-B₃)

3.2. Fourier Transmission Infrared Region(FT-IR) Spectroscopy of the compounds (B₁-B₃).

The spectroscopic results of the prepared compounds (B_1 - B_3) show clear differences. These differences indicate the occurrence of Schiff base reaction.

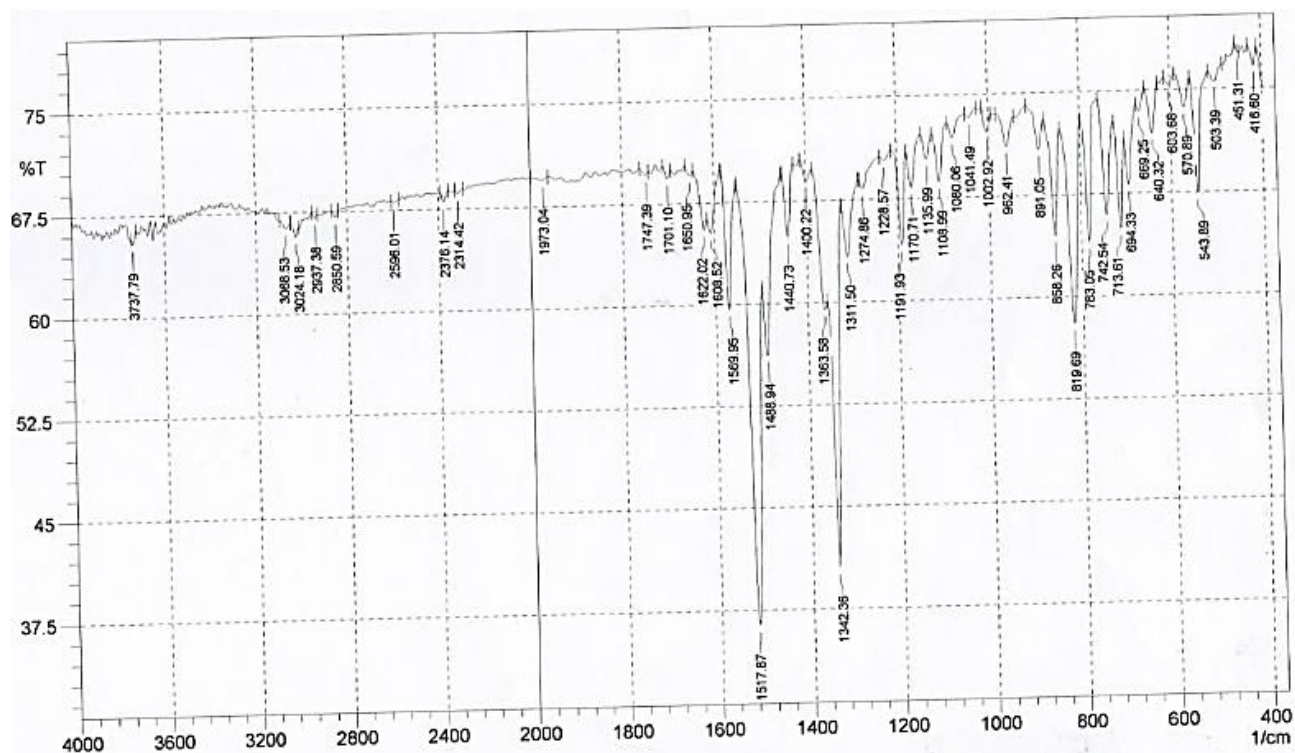


Figure (3): FT-IR spectrum of compound (B_1)

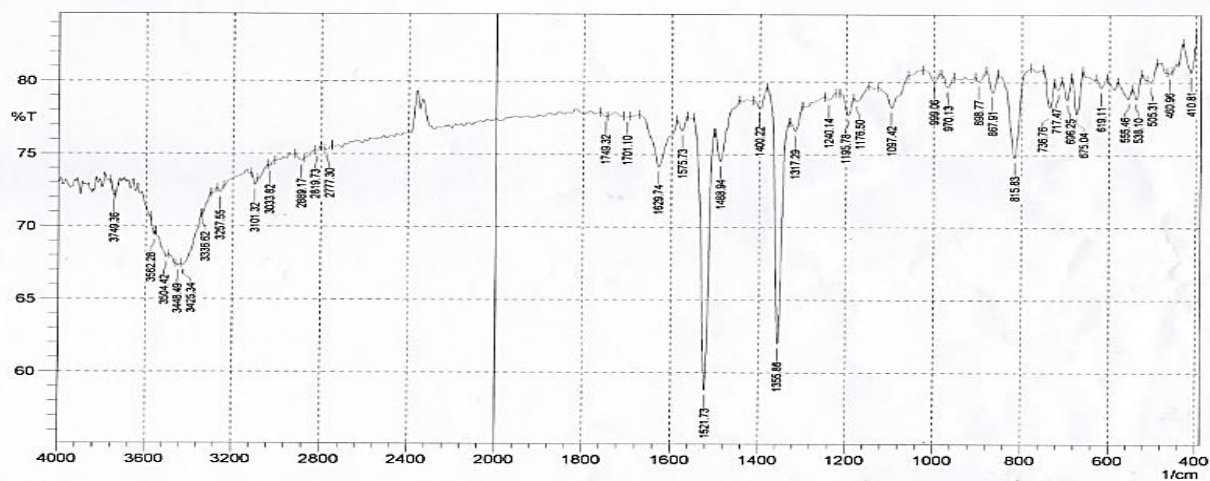


Figure (4): FT-IR spectrum of compound (B_2)

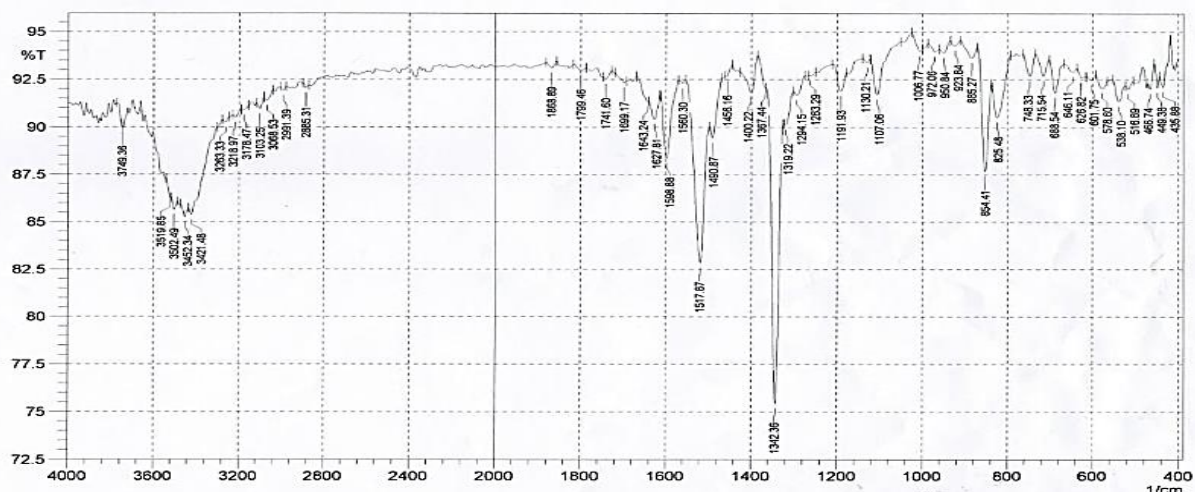


Figure (5): FT-IR spectrum of compound (B₃).

The FT-IR spectrum shows the main difference in the groups in terms of the absence of the carbonyl band and the presence of other bands indicating the presence of imine group $\nu(\text{C}=\text{N})$ at $1662\text{-}1606\text{ cm}^{-1}$. According to Figures (3-5), the band appeared at $1701\text{-}1708\text{ cm}^{-1}$ for $\nu(\text{C}=\text{O})$ of the carbonyl group, the band of $\nu(\text{O-H})$ appeared at $3438\text{-}3288\text{ cm}^{-1}$, the band of aromatic $\nu(\text{C}=\text{C})$ appeared at $1591\text{-}1510\text{ cm}^{-1}$ [15-17].

3.3. Atomic Force Microscope (AFM)

Three-dimensional (3D) AFM measurements showed the carbon steel surface immersed in 3.5% NaCl solution, in the absence of inhibitors. Then the Peaksto-valley value of AFM image analysis was performed to obtain the mean roughness S_a (the mean deviation for all coils). Define the roughness of points from the mean line along the evaluation). While the root-square roughness, S_q (mean measured height deviations taken within the evaluation length and measured from the mean line), and peak height S_y values (the largest single peak height in five adjacent sampling heights). Figure (6) (1 and 2) shows the corroded metal surface with few pits in the absence of inhibitors. (S_a), (S_q), (S_y) for the carbon steel surface were 2.79 nm, 4.11 nm, and 42.8 nm, respectively, the average roughness was reduced as shown in Figures (7, 8, 9) (3 and 14).), which appears on the surface of the steel after immersion in 3.5% NaCl in the presence of an inhibitor. (S_a), (S_q), (S_y) had higher values and were lower in the presence of the inhibitor than in the absence of the inhibitor. These parameters indicated that the surface was smoother. The smoothness of the surface was due to the formation of an integrated protective layer of the compound inhibitor (B1-B3) on the metal surface, which prevented the corrosion of carbon steel [18].

Table 1-Average roughness (Ra), root mean square roughness (Rq) and mean grain size(Ry) for carbon steel by compounds(B1-B3) film in the absence and presence of the inhibitor.

Compounds	Ra (nm)	RMS (nm)	Mean grain size (nm)
Carbon steel	2.79	4.11	42.8
B ₁ without corrosion	47.5	55	190
B ₁ with corrosion carbon steel	44.6	51.5	178
B ₂ without corrosion	47.5	56.6	234
B ₂ with corrosion carbon steel	31.	36.5	141
B ₃ without corrosion	29.1	34.5	138
B ₃ with corrosion carbon steel	30.2	38.7	154

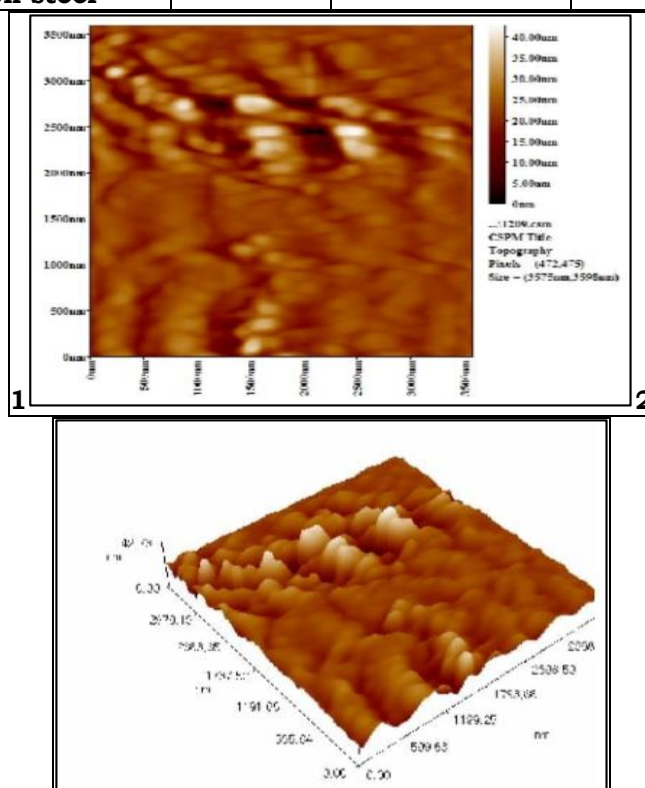
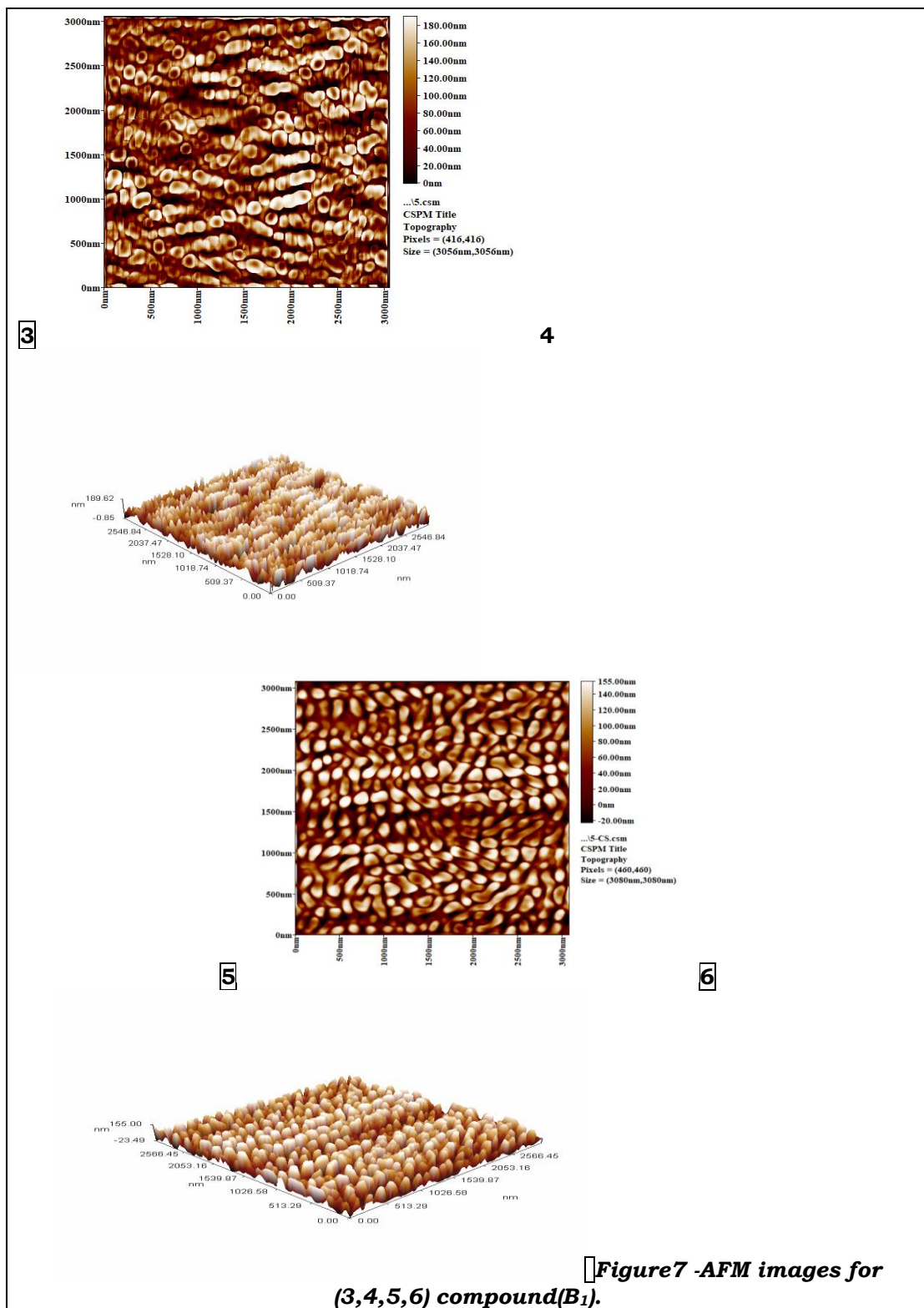
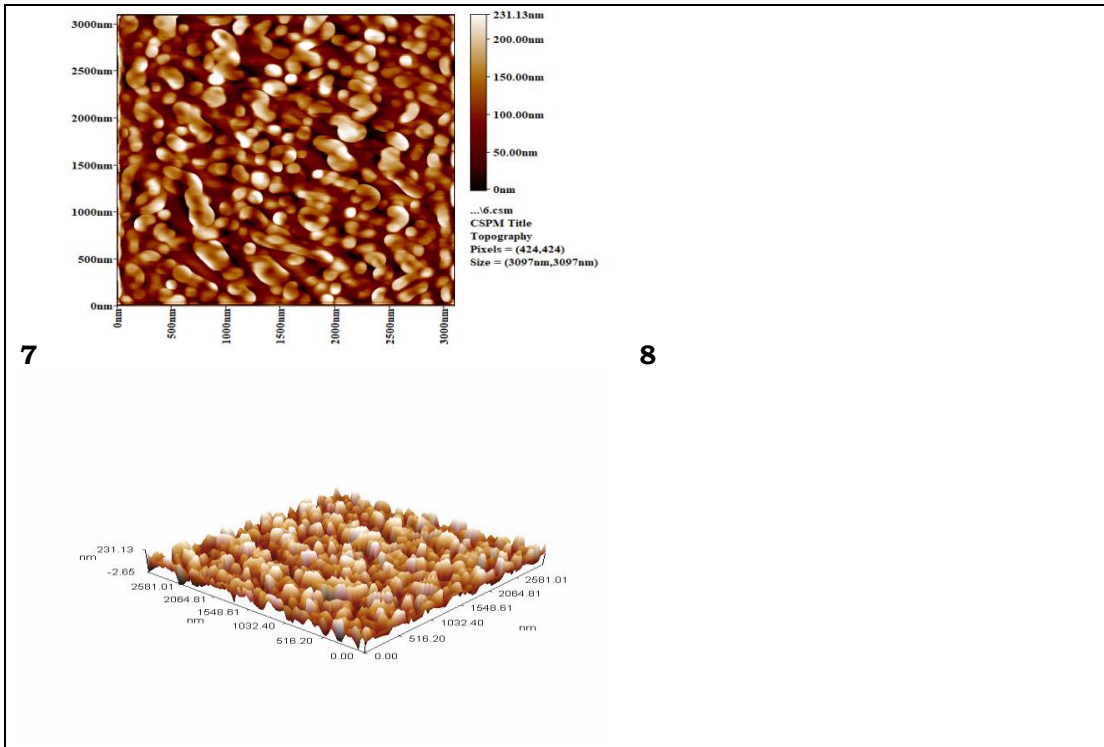


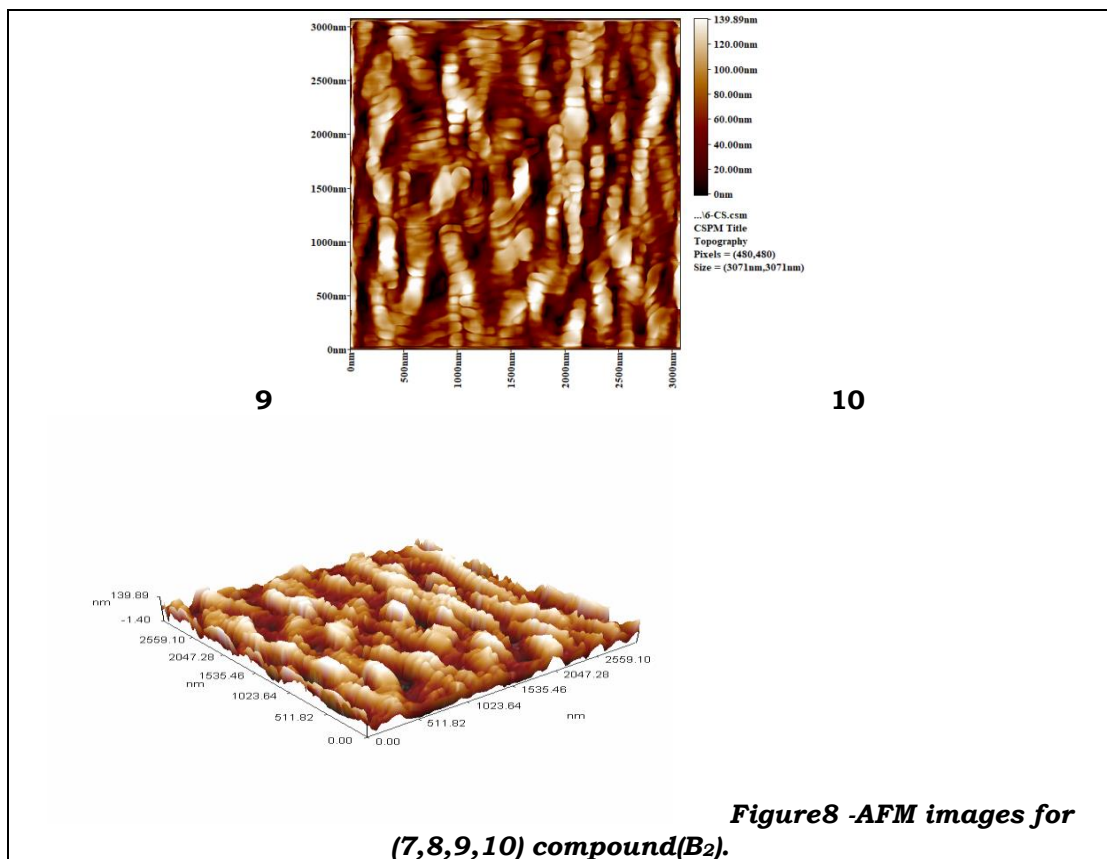
Figure6-AFM images for (1,2) carbon steel.

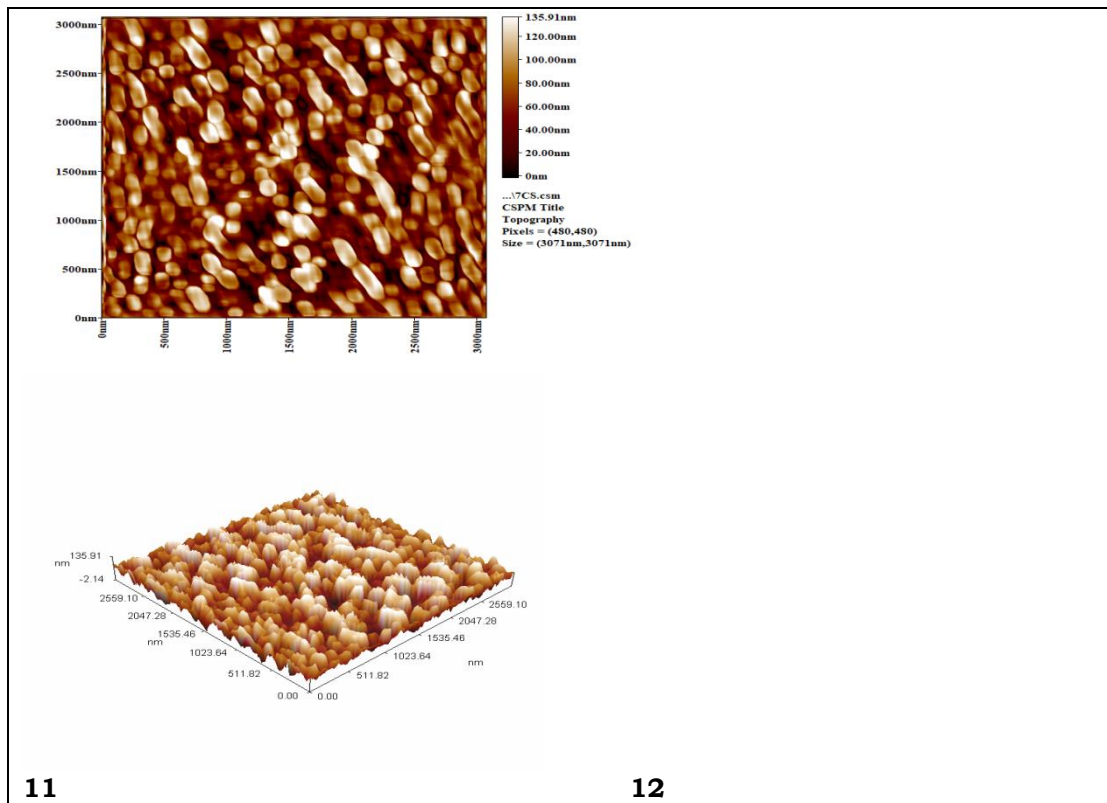




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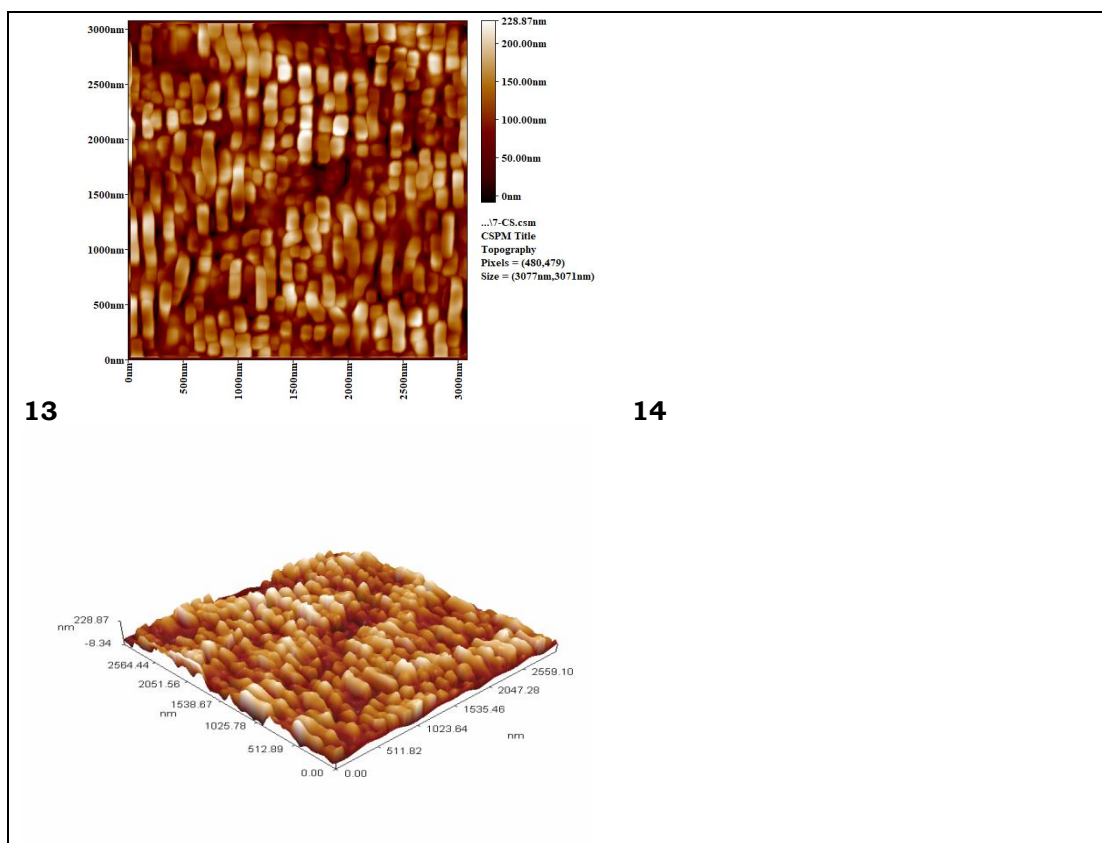


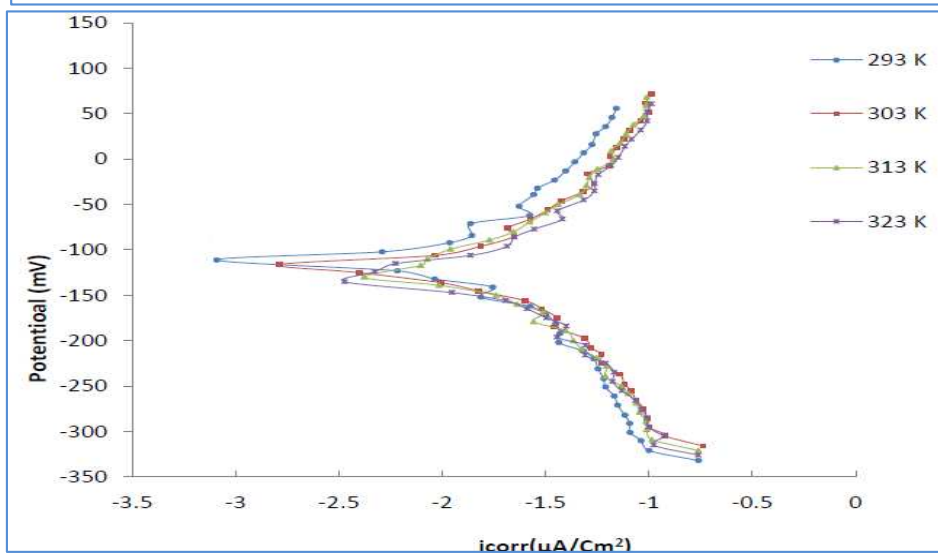
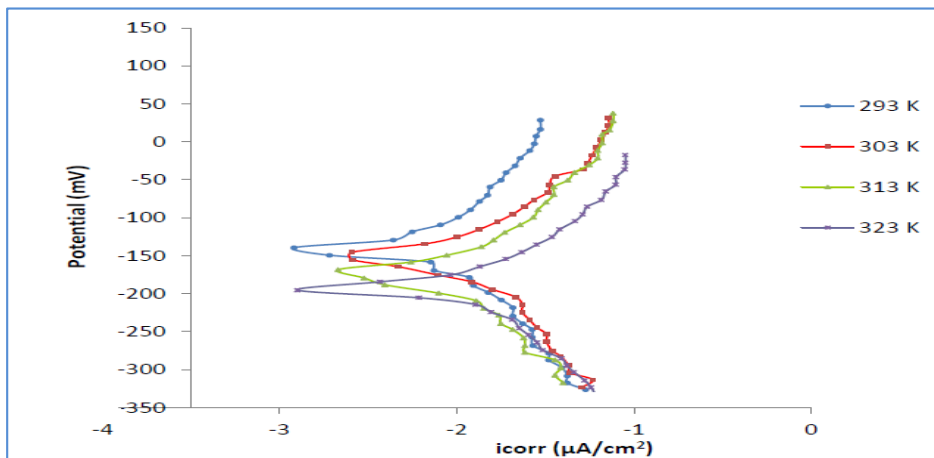
Figure9 -AFM images for (11,12,13,14) compound (B₃).

3.4. Corrosion parameters

The corrosion parameters were determined from the data in Tables (2) and Figure (10). The corrosion potential (E_{corr}) and corrosion current density (i_{corr}) were determined by extrapolation of the cathodic and anodic Tafel lines to the point of intersection for uncoated and coated C.S with Schiff base in presence and absence of inhibitors in NaCl 3.5% solution [19]. The cathodic (β_c) and anodic (β_a) Tafel slopes were calculated from the same Figure (10). Table (2) show the data of the corrosion potential E_{corr} (mV), corrosion current density i_{corr} ($\mu\text{A}/\text{cm}^2$), anodic and cathodic Tafel slopes β_a and β_c (mV/Dec) and protection efficiencies (PE%). The values of PE% was determined from the following equation [20]:

$$\text{PE}\% = [I \text{ corr (uncoated)} - I \text{ corr (coated)}] / [I \text{ corr (uncoated)}] * 100 \dots(5)$$

Where i_{0corr} and i_{corr} the uncoated and coated corrosion current densities respectively, obtained by the extrapolation of cathodic Tafel lines to corrosion potential.



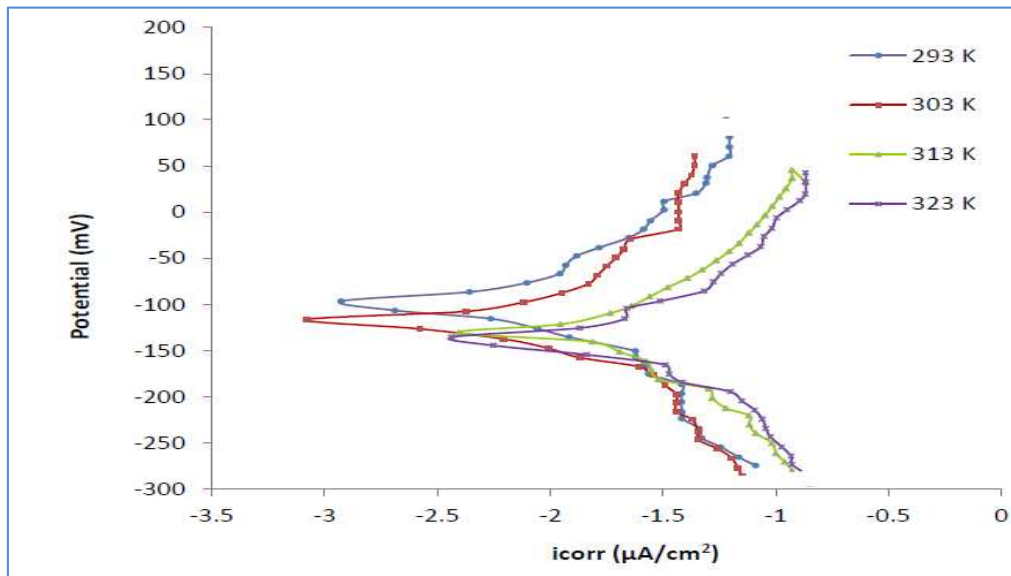


Figure 10: Polarization curves for corrosion of a) uncoated C.S, b) coated C.S with compound(B₁), c) coated C.S with compound(B₂),d) coated C.S with compound(B₃).

Table 2.: Corrosion data of carbon steel in 3.5% NaCl in blank and presence of various concentrations at a temperature of (293 -323)K.

Coating	T(K)	E _{corr} (mV)	icorr (μA/cm ²)	-β _c (mV/sec)	β _a (mV/sec)	PE%	
Blank	293	621.0	121.7	146.9	82.1		
	308	708.0	164.5	26.3	73.8		
	318	785.7	191.1	102.0	31.2		
	323	667.2	195.2	129.9	82.3		
Compound (B₁)	293	88.2	27.44	224.0	102.7	77.43	0.775
	303	154.1	41.84	454.2	106.8	74.565	0.746
	313	174.0	55.16	114.1	79.0	71.136	0.714
	323	202.2	55.86	148.2	88.1	71.383	0.714
Compound (B₂)	293	120.6	24.6	231.2	79.0	79.786	0.798
	303	126.8	36.09	426.4	91.4	78.061	0.781

	8							
	31	146.8	45.52	106.0	59.4		76.188	0.762
	8	3						
	32	150.6	50.23	102.6	86.8		74.267	0.743
	3	5						
Compound (B₃)	29		20.48	358.6	55.7		83.172	0.832
	3	92.93						
	30		32.53	177.5	78.6		80.225	0.802
	3	130.8						
	31		42.01	132.4	98.5		78.017	0.780
	3	144.65						
	32		44.08	222.8	92.9		77.418	0.774
	3	150.88						

The data of the Table (2) show that the corrosion potential (E_{corr}) and corrosion current density (i_{corr}) increased with temperature increasing. The corrosion current density decreased for coated C.S than bare C.S, the current density was decreased. The Tafel plots showed the corrosion potential (E_{corr}) of the coated C.S (working electrode) shifted into higher position (noble direction) compared to that of uncoated C.S. This indicates that the corrosion protection act as anodic protection [21,20]. The cathodic (β_c) and anodic (β_a) Tafel slopes obtained from the slopes of the cathodic and anodic Tafel regions show in Tables (2). The results of anodic and cathodic. Tafel slopes (β_a and β_c) varied at all temperatures These results refer to the variation of the rate determining step from charge transfer process to electrochemical desorption and chemical deposition in the cathodic reaction and also to the rate demining step variation in the metal dissolution reaction [23].

The data of the Table (2) show that the The PE% values decreased with an increasing of temperature that can be explained by the fact the thickness of the boundary layer decreased with increasing of the temperature, and that due to increase the ability of the desorption of some Schiff base[24].

3.5. Temperature effect on the corrosion of C.S

The corrosion process for uncoated and coated C.S in of NaCl 3.5% solution in a temperature range (293-323) K was studied by using similar Arrhenius equation [25].

$$i_{corr} = A \exp(-E_a/RT) \quad \text{.....(6)}$$

Equation converted into logarithmic form as follows:

$$\text{Log}(i_{corr}) = \text{Log} A - E_a/2.303 RT \quad \text{.... (7)}$$

While the transition state equation expressed as follows [26]:

$$\text{Log}(i_{corr}/T) = \text{Log}(R/Nh) + \Delta S^*/2.303 R - \Delta H^*/2.303 RT \quad \text{.... (8)}$$

Where E_a : activation energy, A : pre-exponential factor, R : gas constant (8.314 J/K. mol), i_{corr} : corrosion current density, and T : absolute temperature (K), N : the Avogadro's number ($6.022 \times 10^{23} \text{ mol}^{-1}$), h : Plank's constant ($6.626 \times 10^{-34} \text{ J.s}$), ΔS^* : the entropy of activation, ΔH^* : the enthalpy of activation. Figures (11) show Arrhenius plots for the corrosion of both uncoated and coated C.S in

NaCl 3.5% solution. The values of E_a and A derived from the slopes and intercept of the plots respectively. Figures (12) show the plots of $\log(i_{corr}/T)$ versus $1/T$ by which the enthalpy change of activation (ΔH^*) and entropy change of activation (ΔS^*) obtained from the slopes and intercepts respectively. The activation Gibbs free energy (ΔG^*) determined by using the following equation [27].

$$\Delta G^* = \Delta H^* - T\Delta S^* \quad \dots\dots$$

(9)

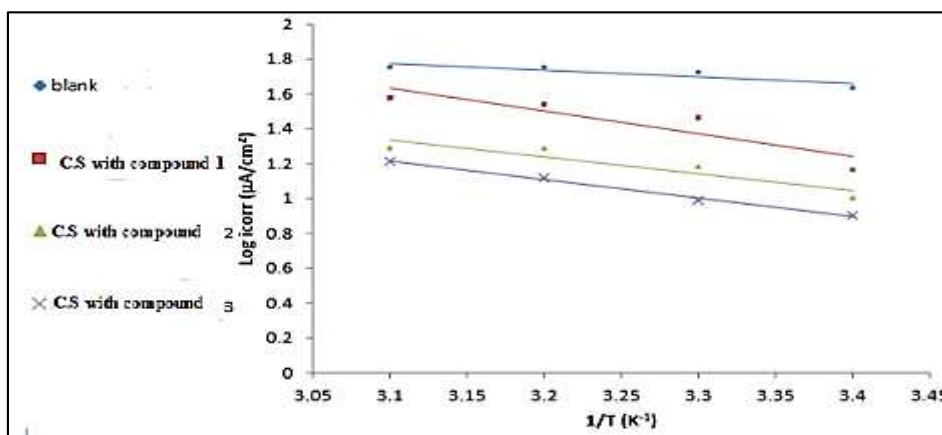


Figure 11-Plot of $\log i_{corr}$ vs. $1/T$ for the compounds(B1-B3) in the absence and presence of the inhibitors in 3.5% NaCl solution.

Table 3-Transition state thermodynamic parameters for the corrosion of the uncoated and coated C.S with a compounds in the absence and presence of the inhibitor in 3.5% NaCl solution.

Coating.	E_a KJ mol ⁻¹	A Molecules cm ⁻² S ⁻¹	R^2
Blank	17.853	$1.06 \cdot 10^{29}$	0.78
Compound(B ₁)	27.562	$1.14 \cdot 10^{30}$	0.818
Compound(B ₂)	24.304	$2.74 \cdot 10^{29}$	0.859
Compound(B ₃)	28.386	$1.19 \cdot 10^{30}$	0.994

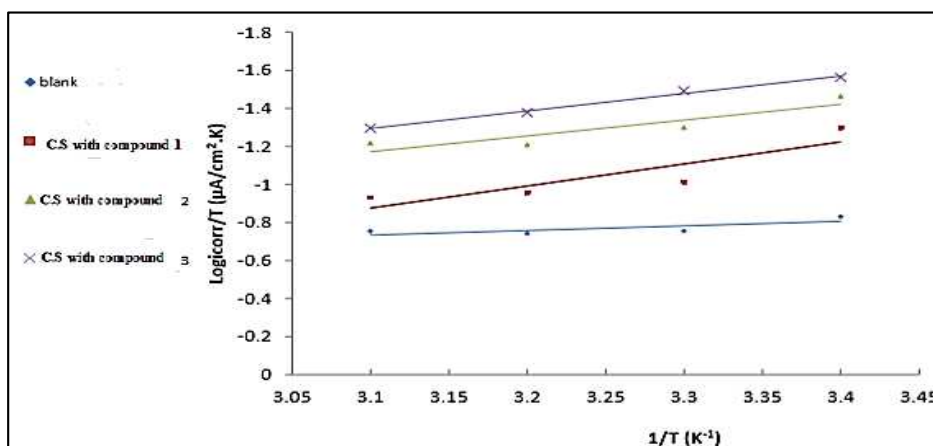


Figure 12-Plot of $\log i_{\text{corr}}/T$ vs. $1/T$ for the compounds (B_1 - B_3) in the absence and presence of the inhibitors in 3.5% NaCl solution.

Table 4-Transition state thermodynamic parameters at different temperatures for the corrosion of the uncoated and coated C.S with a compounds film in the absence and presence of the inhibitor in 3.5% NaCl solution

Coating.	$\Delta G/ \text{kJ mol}^{-1}$				ΔH^* kJ mol^{-1}	$-\Delta S^*$ $\text{J mol}^{-1} \cdot \text{K}^{-1}$	R^2
	298K	303K	313K	323K			
Blank	61.186	62.726	64.266	66.75	15.294	0.154	0.80
Compound(B_1)	64.638	65.968	67.298	68.407	23.004	0.133	0.783
Compound(B_2)	64.954	66.404	67.854	68.279	21.744	0.143	0.821
Compound(B_3)	65.462	66.792	68.122	70.032	25.828	0.133	0.993

4. Conclusion

The corrosion current density (i_{corr}) and corrosion potential (E_{corr}) increased with increasing temperature. The corrosion current density (i_{corr}) decreased after coating the C.S with the inhibitors in the absence and presence of the inhibitors. Protection efficiency (PE%) was decreased with increasing temperature for the uncoated and coated C.S in the absence and presence of the inhibitors. The compound B_3 coated C.S had a higher protection efficiency (PE%) than the compound (B_1, B_2) coated C.S. AFM and SEM analysis showed that the protection of C.S occurred due to the formation of a protective film on the metal surface.

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