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Corrosion inhibition of 1018 carbon steel in hydrochloric acid using Schiff base compounds

I. A. Zaafarany

Chemistry Department, Faculty of Sciences, Umm Al-Qura University, Makkah,
Saudi Arabia. E-mail: ishaq_zaafarany@yahoo.com

Abstract

The inhibition effect of some Schiff base compounds toward the corrosion of 1018 carbon steel in hydrochloric acid solution was studied using weight loss, galvanostatic polarization, and electrochemical impedance spectroscopy measurements. The percentage inhibition efficiency increased with increasing inhibitor concentration and with decreasing temperature. The polarization measurements indicated that the inhibitors are of mixed type and inhibit corrosion by adsorption on the steel surface. The adsorption process is described by the Freundlich adsorption isotherm. The activation energy and some activation thermodynamic parameters were calculated and explained. From the impedance data it is found that the corrosion of carbon steel is controlled by the charge transfer process at all concentrations of inhibitors.

Keywords: *Schiff base compounds, 1018 carbon steel, adsorption, corrosion inhibitors.*

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1. Introduction

Carbon steel is widely used in many industrial applications. In most industrial processes, the acidic solutions are commonly used for the pickling, industrial acid cleaning, and acid descaling, oil well acidifying, *etc*. [1 – 3]. Because of the general corrosivity of acid solutions, inhibitors are commonly used to reduce the corrosive attack on the steel surface [4]. Several studies on using organic compounds to inhibit the corrosion of carbon steel in acidic solutions have been reported [5 – 15]. It has been accepted that the corrosion inhibition by organic compounds process results from its adsorption at the metal–solution interface [16]. The adsorption process is dependent on the chemical structure of the molecule, chemical composition of the solution, nature of the metal surface and the electrochemical potential of the metal – solution interface.

The aim of the present work is to study the effect of some Schiff base compounds as inhibitors on the corrosion of carbon steel (Type 1018) in 1 M HCl using weight loss, galvanostatic polarization, and electrochemical impedance measurements. The effect of temperature on the corrosion of carbon steel in 1 M HCl containing the inhibitors used was also studied and some thermodynamic parameters were computed.

2. Experimental methods

1018 carbon steel used in this study has the chemical composition C 0.2%, Mn 0.6%, P 0.04 %, Si 0.003% and the remainder is iron. Weight loss measurements were performed using coupons of the dimensions $3 \times 3 \times 0.1$ cm^3. The steel coupons were left hanged in the test solution (1 M HCl) for 150 minutes at 25 ± 1°C before recording the loss of their weight. Weight loss measurements were carried out as described elsewhere [17]. The percentage inhibition efficiency (*IE*) of the Schiff base compounds was calculated from weight loss measurements using the following equation:

$$\mathrm{IE} = \left[1 - \frac{W_{add}}{W_{free}} \right] \cdot 100 \qquad (1)$$

where, W_{add} and W_{free} are the weight loss of the metal in presence and absence of the inhibitor.

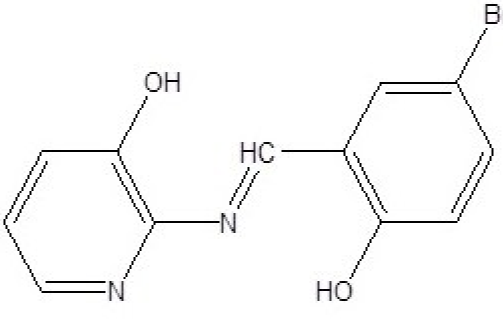
For galvanostatic polarization, a cylindrical rod embedded in araldite with exposed surface area of 0.44 cm^2 was used. The electrodes were polished with different grades emery papers, degreased with acetone and finally rinsed by distilled water, before immersing in the test solution. Galvanostatic polarization studies were carried out using a (PS remote) potentiostat with Zm PS6 software for calculation of corrosion parameters. A three compartment cell with a saturated calomel reference electrode was used. The inhibition efficiency *IE* was calculated using the following equation:

$$IE = \left[1 - \frac{I_{corr.add}}{I_{corr.free}} \right] \cdot 100 \qquad (2)$$

where $I_{corr.add}$ and $I_{corr.free}$ are the corrosion rates in free and inhibited acid, respectively.

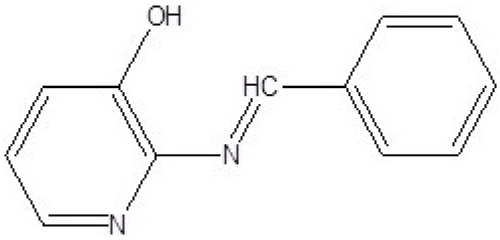
The chemical structures of the Schiff base compounds are:

Compound 1:



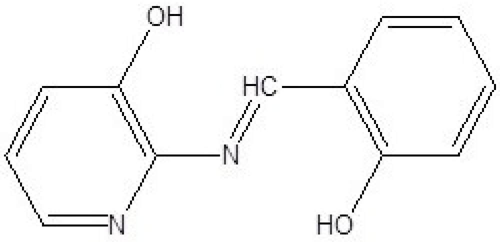
2-(5-Bromo-2-hydroxybenzlideneamino)pyridine-3-ol

Compound II:



2-(Benzylideneamino)pyridine-3-ol

Compound III:



2-(2-Hyroxybenzylideneamino)pyridine-3-ol

3. Results and discussion

3.1. Weight loss measurements

Figure 1 represents the relation between time and weight loss of 1018 carbon steel coupons in 1 M HCl solution in absence and presence of different concentrations of compound III as an example of the tested Schiff base compounds. Similar curves were also obtained for other two compounds (not shown). It is obvious that the weight loss of 1018 carbon steel decreases and is much lower than that obtained in the blank solution. The linearity obtained indicates the absence of insoluble surface film during corrosion and that the inhibitors were first adsorbed onto the surface and thereafter impede the corrosion process [18]. The calculated values of inhibition efficiency (*IE*) and surface coverage θ from the weight loss measurements are listed in Table 1. It is obvious that the values of *IE* increase with the inhibitor concentration.

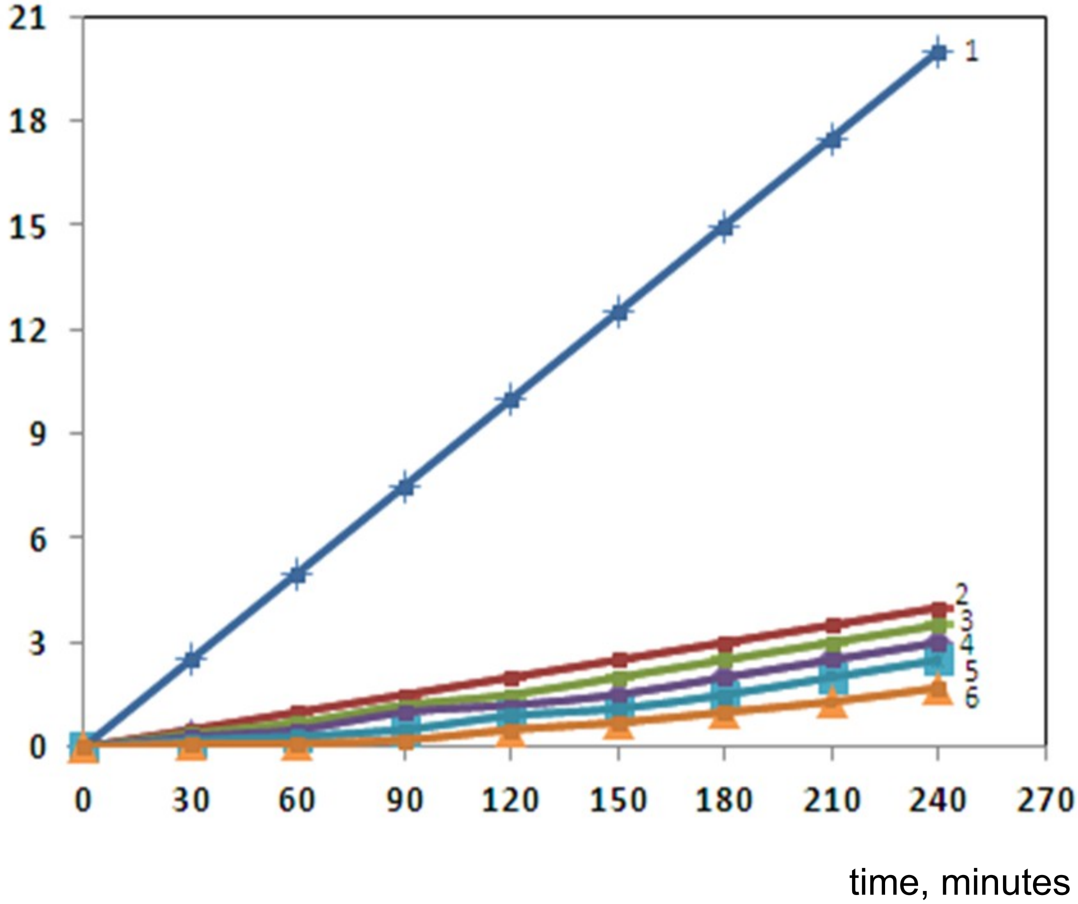


Fig. 1. Weight loss – time curves for C-steel in 1 M HCl solution in absence and presence of different concentrations of compound I: *1*, 0.00 ppm; *2*, 100 ppm; *3*, 200 ppm; *4*, 300 ppm; *5*, 400 ppm; *6*, 500 ppm.

Table 1. Effect of inhibitors on the corrosion of C-steel in 1.0 M HCl solution using weight loss measurements at 30°C.

Concentrations	**R_{corr}, mg cm^{-2} min^{-1}**	**% *IE***	**θ**
	1 M HCl + compound I		
0.00 ppm compound I	0.836	–	–
100 ppm compound I	0.224	73.20	0.732
200 ppm compound I	0.0146	82.53	0.825
300 ppm compound I	0.0098	88.27	0.883
400 ppm compound I	0.0076	90.90	0.909
500 ppm compound I	0.0052	93.77	0.937

Concentrations	R_{corr}, mg cm^{-2} min^{-1}	**% *IE***	**θ**
	1 M HCl + compound II		
100 ppm compound II	0.0196	76.55	0.765
200 ppm compound II	0.0108	87.08	0.871
300 ppm compound II	0.0078	90.66	0.901
400 ppm compound II	0.0058	93.06	0.931
500 ppm compound II	0.0036	95.69	0.957
	1 M HCl + compound III		
100 ppm compound III	0.0165	80.26	0.803
200 ppm compound III	0.0088	89.47	0.895
300 ppm compound III	0.0064	92.34	0.923
400 ppm compound III	0.0035	95.81	0.958
500 ppm compound II	0.0028	96.65	0.966

This behavior could be attributed to the increased surface coverage θ due to the increase of the number of adsorbed molecules at the metal surface. At one and the same inhibitors concentration, the percentage of inhibition efficiency decreases in the following sequences:

compound III > compound II > compound I.

This behavior will be discussed later.

3.2. Adsorption isotherm

The adsorption of Schiff base compounds on the surface of steel electrode is regarded as substitutional adsorption process between the organic compound in the aqueous phase (Org) and the water molecules adsorbed on the steel surface $(H_2O)_{ads}$ [19].

$$\mathrm{Org}_{(sol)} + X(\mathrm{H_2O})_{(ads)} \rightleftharpoons \mathrm{Org}_{(ads)} + X\mathrm{H_2O}_{(sol)}, \tag{3}$$

where X is the size ratio, that is, the number of water molecules replaced by one organic molecule. The values of surface coverage θ for different concentrations of the studied Schiff base compounds at 30°C have been used to explain the best isotherm to determine the adsorption process. Attempts were made to fit θ values to various isotherms including Frumkin, Temkin, Freundlich and Langmuir adsorption isotherms. By far, the best fit was obtained with Freundlich isotherm according to the following equation [20]:

$$\theta = KC^{n} \tag{4}$$

or in the logarithmic form

$$\log \theta = \log K + n \log C. \tag{5}$$

Fig. 2 represents the relation between log (θ) and log C_{Inh}. Straight lines relationship were obtained for three compounds studied suggesting that the adsorption of Schiff base compounds on the carbon steel surface obeys the Freundlich isotherm. The values of the equilibrium constant of adsorption were calculated from the intercept and equal to 5.02, 5.62 and 6.31 for compounds I, II and III, respectively.

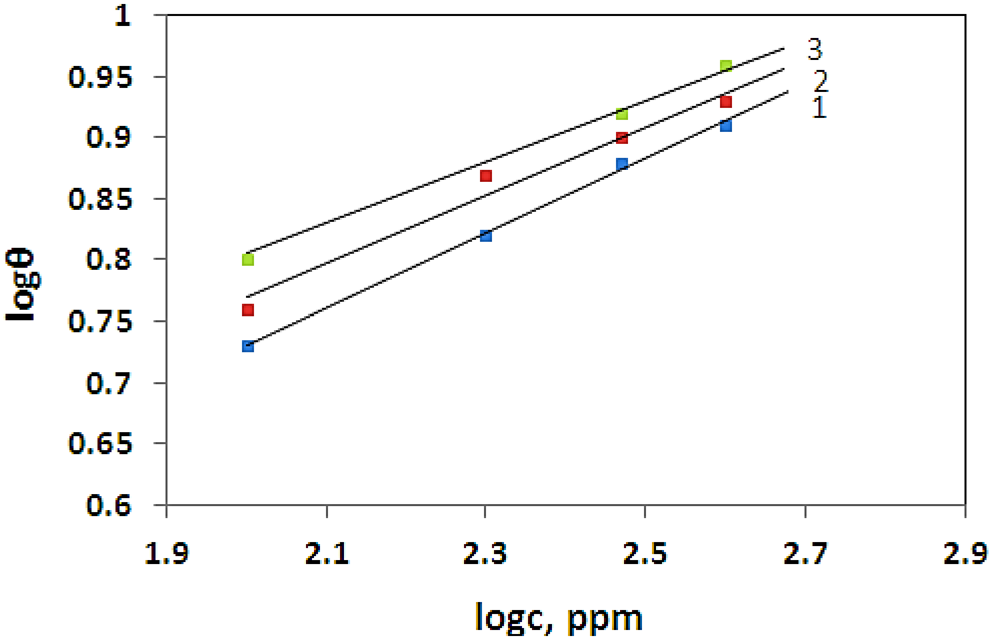


Fig. 2. Freundlich isotherm for C-steel electrode in 1.0 M HCl solution in the presence of additives. *1*, compound I; *2*, compound II; *3*, compound III.

The values of $\Delta G^0_{\mathrm{ads}}$ were calculated from the following equation:

$$K = 1/55.5\exp(-\Delta G^0_{\mathrm{ads}}/RT) \qquad (6)$$

and equal to –18.87, –23.95 and –30.17 kJ mol^{-1} for compounds I, II and III, respectively. The standard free energy change of adsorption is associated with water adsorption/desorption equilibrium which forms an important part in the overall free energy changes of adsorption. The negative values of $\Delta G^0_{\mathrm{ads}}$ obtained have indicated that the adsorption process of these compounds on the steel surface is spontaneous one.

3.3. Effect of temperature

The effect of rising temperature on the corrosion rate of 1018 carbon steel in 1 M HCl solution in absence and presence of 500 ppm of three compounds of Schiff base was studied by weight loss measurements over a temperature range from 30-60°C. Similar curves to Fig. 1 were obtained (not shown). Values of corrosion rate R_{corr} and inhibition efficiency *IE* obtained at different temperatures are listed in Table 2. Inspection of Table 2 reveals that, as the temperature increases, the rate of corrosion increases and hence the inhibition efficiency of the additives decreases. This is due to the desorption is aided by increasing the temperature. This behavior proves that the adsorption of inhibitors on C-steel surface occurs through physical adsorption.

The activation energy (E_a) of the corrosion process was calculated using Arrhenius equation [21]:

$$R_{corr} = Ae^{-E_a^*/RT} \tag{7}$$

And in the logarithmic form

$$\log R_{corr} = \log A + \frac{E_a}{2.303\ RT}, \tag{8}$$

where R_{corr} is the rate of corrosion from weight loss, A is Arrhenius constant, R is the gas constant and T is the absolute temperature.

Figure 3 represents Arrehenius plots (log R_{corr} *vs*. $1/T$) for uninhibited and inhibited 1 M HCl containing 500 ppm of the studied compounds. The values of E_a can be obtained from the slope of the straight lines were found to be 16.253 kJ mol^{-1} in 1 M HCl and 20.205, 22.043 and 24.655 kJ mol^{-1} in presence of compound I, II and III, respectively. It is obvious that the values of E_a^* increase in the presence of the inhibitors. This was attributed to an appreciable decrease in the adsorption process of the inhibitors on the steel surface with increase of temperature and corresponding increase in the reaction rate because of the greater area of the metal that is exposed to HCl solution [22].

Table 2. Effect of rising temperature on the corrosion rate and inhibition efficiency for the corrosion of C-steel in 1.0 M HCl solution in absence and presence of 500 ppm of inhibitor using weight loss measurements.

Inhibitors	***T*, °C**	**R_{corr}, mg cm^{-2} min^{-1}**	**% *IE***
Free 1.0 M HCl			
	30	0.0836	–
	40	0.1125	–
	50	0.1354	–
	60	0.1568	–
1 M HCl + 500 ppm compound I			
	30	0.0052	93.77
	40	0.0114	89.86
	50	0.0266	83.03
	60	0.0314	79.74
1 M HCl + 500 ppm compound II			
	30	0.0036	95.69
	40	0.0092	91.82

Inhibitors	***T*, °C**	***R*corr, mg cm^{-2} min^{-1}**	**% *IE***
	50	0.0195	85.59
	60	0.0282	82.01
1 M HCl + 500 ppm compound III			
	30	0.0028	96.65
	40	0.0083	92.62
	50	0.0163	87.96
	60	0.0249	84.11

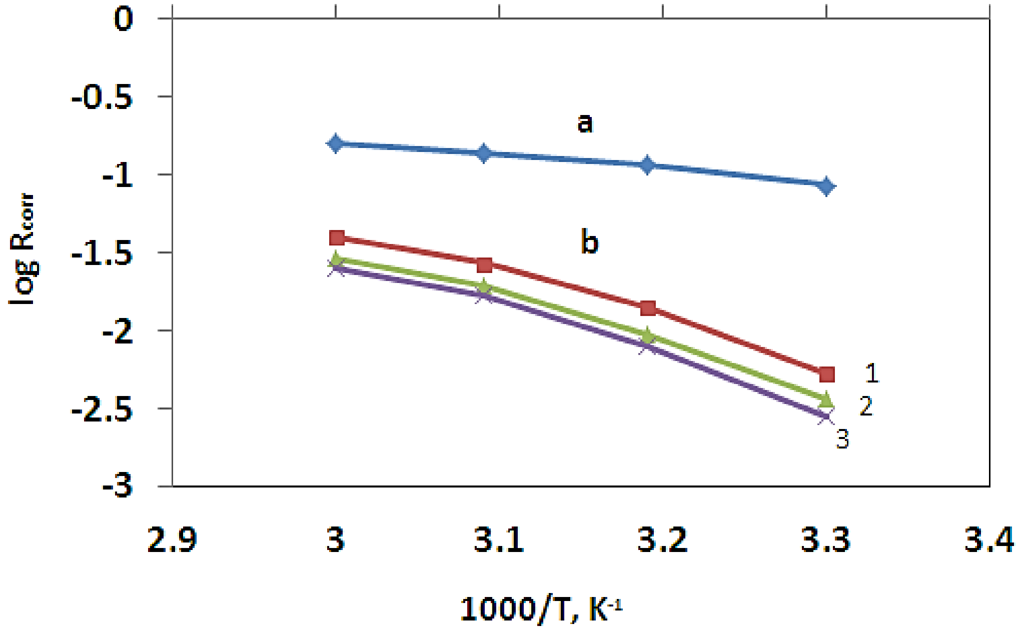


Fig. 3. log R$_{\text{corr}}$ – 1000/*T* curves for C-steel electrode in 1M HCl solution in the absence and in the presence of additives. *a*, 1 M HCl; *b*, 1 M HCl + 500 ppm of inhibitor. *1*, compound I; *2*, compound II; *3*, compound III.

The activation of some thermodynamic parameters, *e.g.*, the enthalpy of activation ΔH^* and the entropy of activation S^* for the corrosion of carbon steel in 1.0 M HCl solutions in the absence and presence of 500 ppm of Schiff base compounds were calculated from transition state equation [21]:

$$R_{\text{corr}} = RT/Nh \exp(S^*/R) \exp(-\Delta H^*/RT) \qquad (9)$$

where *A* is the frequency factor, *h* is the Plank constant, *N* is Avogadro number and *R* is the universal gas constant.

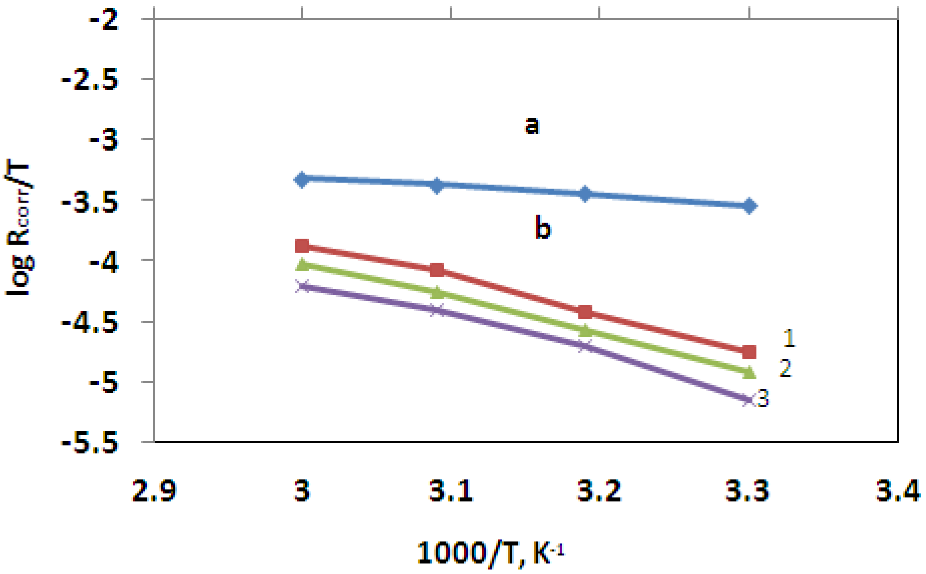


Fig. 4. Relation between log R_{corr} and $1/T$ of C-steel electrode in: *a*, 1 M HCl; *b*, 1 M HCl + 500 ppm of inhibitor. *1*, compound I; *2*, compound II; 3, compound III.

Figure 4 shows a plot of log (R_{corr}/T) *vs*. $1/T$ for uninhibited carbon steel electrode in 1 M HCl solution in the absence and presence of 500 ppm of the studied compounds. The above plots give straight lines with a slop of ($-\Delta H^*/2.303R$) and an intercept of [log (R/Nh) + $\Delta S^*/2.303R$]. The values of ΔH^* obtained from the slope of the straight line equal to 35.28 kJ mol^{-1} in the case of free inhibited solution, and equal to 47.85, 54.63 and 59.75 kJ mol^{-1} in presence of compound I, II and III, respectively. The data obtained from the experimental results indicate that the values of ΔH^* are positive. Hence the process is endothermic. The values of ΔH^* are different for studied compounds which means that their structure affect the strength of its adsorption on the metal surface. The values of ΔS^* obtained from the intercept of the straight line are equal to –349.65 J K^{-1} mol^{-1} in the case of free inhibited solution, and equal to 425.75, 465.53 and 498.82 J K^{-1} mol^{-1} in the presence of compound I, II and III, respectively. The negative values of ΔS^* in the absence and presence of the inhibitors implies that the activated complex is the rate determining step and represents association rather than dissociation. It also reveals that an increase in the order takes place in going from reactants to the activated complex [22]. The inhibition efficiency of Schiff base compounds as gathered from the increase in E_a^* and ΔH^* values and decrease in ΔS^* decreases in the following sequence:

compound III > compound II > compound I

3.4. Galvanostatic polarization technique

Figure 5 shows the anodic and cathodic polarization curves (Tafel plots) for C-steel in 1.0 M HCl solution devoid of and containing different concentrations of compound III at 25°C as an example. Similar curves were obtained in presence of the other two compounds (not shown). Some corrosion parameters, *e.g.*, anodic (b_a) cathodic (b_c) Tafel slopes, corrosion potential (E_{corr}.), corrosion current density (I_{corr}.) and the percentage inhibition efficiency (% *IE*), were calculated and listed in Table 3.

Inspection of Fig. 5 and Table 3, it is clear that both anodic steel dissolution and cathodic hydrogen evolution reactions were inhibited when investigated inhibitors were added to 1 M HCl and this inhibition was more pronounced with increasing inhibitor concentration. Tafel lines are shifted to more negative and more positive potentials with respect to the blank curve by increasing the concentration of the investigated inhibitors.

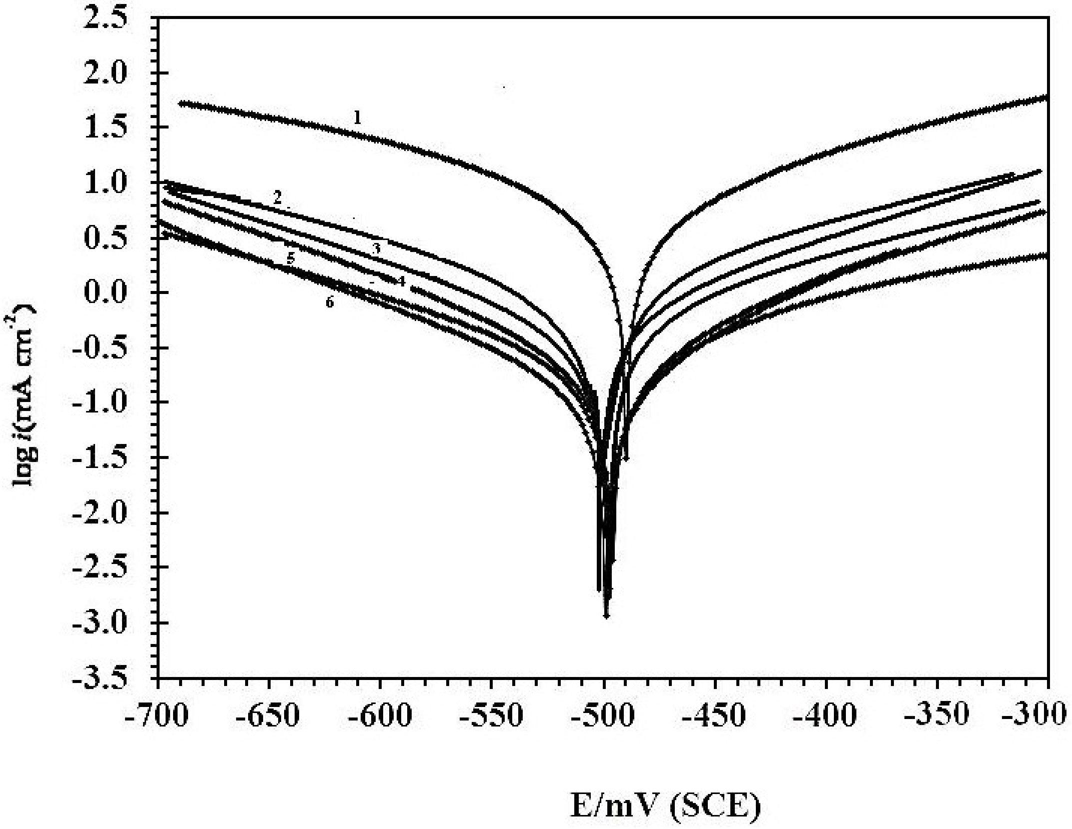


Fig. 5. Galvanostatic polarization curves of C-steel electrode in 1 M HCl containing different concentrations of compound III: *1*, 0.00; *2*, 100; *3*, 200; *4*, 300; *5*, 400; *6*, 500 ppm.

The values of anodic (b_a) and cathodic (b_c) Tafel slopes are increases slightly. This behavior indicates that the Schiff base compounds act as mixed-type inhibitors [23, 24]. The values of corrosion potential (E_{corr}.) is shifted slightly toward negative direction, the corrosion current density (I_{corr}.) decreases, and the percentage inhibition efficiency (% *IE*) increases. This indicates the inhibiting action of Schiff base compounds. The percentage inhibition efficiency decreases in the following order:

compound III > compound II > compound I

Table 3. Corrosion parameters of C-steel electrode in 1 M HCl solution containing different concentrations of inhibitors.

Concentrations	b_a, V decade^{-1}	b_c, V decade^{-1}	$-E_{corr}$, mV (SCE)	I_{corr}, mA. cm^{-2}	% *IE*
1 M HCl + compound I					
0.00 ppm compound I	130	112	505	63.09	–
100 ppm compound I	155	117	512	16.75	73.45
200 ppm compound I	158	119	518	12.34	80.44
300 ppm compound I	157	120	522	10.48	83.38
400 ppm compound I	160	122	520	6.32	89.98
500 ppm compound I	162	125	528	3.86	93.88
1 M HCl + compound II					
100 ppm compound II	145	115	–512	13.54	78.53
200 ppm compound II	150	117	–526	10.22	83.80
300 ppm compound II	152	112	–522	7.38	88.30
400 ppm compound II	145	118	–530	5.12	91.88
500 ppm compound II	156	124	–532	2.88	95.43
1 M HCl + compound III					
100 ppm compound III	155	116	510	11.22	82.21
200 ppm compound III	150	115	512	6.24	90.11
300 ppm compound III	148	118	518	4.18	93.37
400 ppm compound III	152	120	522	2.12	96.63
500 ppm compound II	160	122	525	1.68	97.33

3.5. Electrochemical impedance spectroscopy (EIS)

The corrosion behaviors of C-steel in all the above conditions were also investigated by the EIS method. The equivalent circuit models used to fit the experimental results were as previously reported [25]. Figure 6 shows the complex-plane impedance plots (Nyquist plots) for C-steel in 1 M HCl solution without and with various concentrations of inhibitor at 30°C.

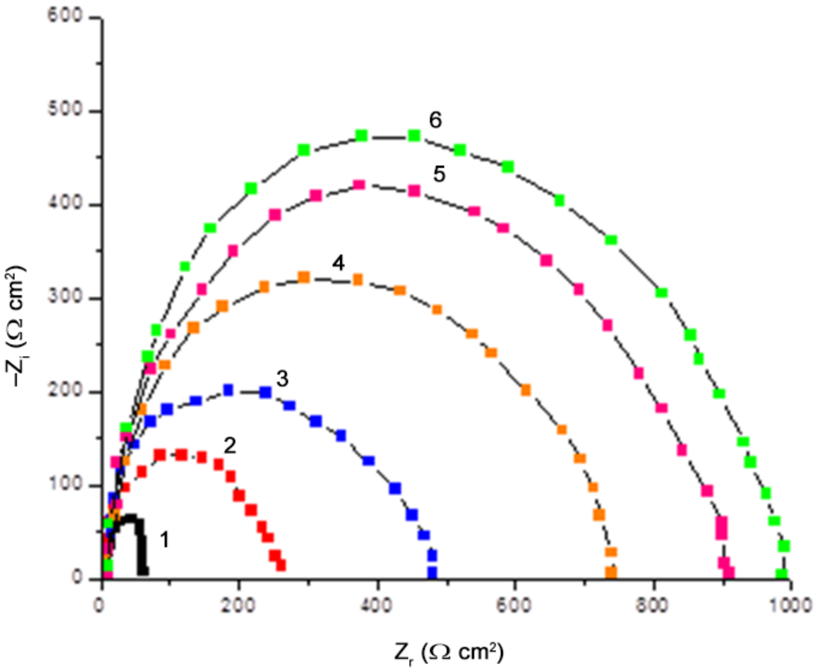


Fig. 6. The Nyquist plots for corrosion of C-steel in 2 M HCl in the absence and presence of different concentrations of compound I at 25°C. *1*, 0.00; *2*,100; *3*, 200; *4*, 300; *5*, 400; *6*, 500 ppm.

As it can be seen from the Fig. 6, the Nyquist plots contain depressed semi-circle with the centre under the real axis, whose size increases with the inhibitor concentration, indicating a charge transfer process mainly controlling the corrosion of carbon steel. Such behaviour, is characteristic for solid electrodes and often refers to a frequency dispersion, has been attributed to roughness and other inhomogeneities of the solid surface [26, 27]. It is apparent, from these plots that the impedance response of C-steel in uninhibited acid solution has significantly changed after the addition of inhibitor compound in the corrosive solutions. This indicated that the impedance of the inhibited substrate has increased with increasing concentration of inhibitor. The characteristic parameters associated to the impedance diagrams (R_{ct} and C_{dl}) and *IE* (%) are given in Table 4. The *IE* (%) was calculated from the following equation [28]:

$$\% IE = \left[\frac{(1/R_{ct})_0 - (1/R_{ct})}{(1/R_{ct})_0} \right] \times 100 \tag{10}$$

where $(R_{\text{ct}})_0$ and R_{ct} are the uninhibited and inhibited charge transfer resistance, respectively.

The percentage inhibition efficiency decreases in the following order:

compound III > compound II > compound I

Table 4. EIS data of C-steel in 2 M HCl and in the absence and presence of different concentrations of investigated inhibitors at 25°C.

Compound	Concentration, ppm	$C_{dl}\times 10^{-3}$, $\mu Fcm^{-2}\ 10^{-4}$	R_{ct}, Ω cm^2	R_s, Ω cm^2	*IE*%
Blank	0 ppm	112.8	61.0	1.18	–
Inhibitor I	100 ppm	31	222	1.20	72.6
	200 ppm	27	318	2.55	80.6
	300 ppm	23	485	1.92	87.4
	400 ppm	21	661	2.51	90.4
	500 ppm	28	763	2.78	91.8
Inhibitor II	100 ppm	36	248	1.21	75.5
	200 ppm	32	448	2.81	86.2
	300 ppm	29	522	1.97	88.3
	400 ppm	26	781	2.79	92.1
	500 ppm	34	1050	2.96	94.2
Inhibitor III	100 ppm	34.3	264.5	1.22	76.5
	200 ppm	28	487.8	2.74	87.6
	300 ppm	24	749.6	1.96	91.3
	400 ppm	22	906	2.72	93.2
	500 ppm	33	993.8	2.94	93.6

The value of *IE* (%) obtained from EIS, galvanostatic polarization and weight loss measurements are in sequence. As it can be seen from Table 4, the R_{ct} values increased with the increasing concentrations of the inhibitor. On the other hand, the values of C_{dl} decreased with an increase in inhibitor concentration. This situation was the result of increase in the surface coverage by the inhibitor, which led to an increase in the inhibition efficiency (Table 4). This decrease in C_{dl}, which can result from a decrease in local dielectric constant and/or an increase in the thickness of the electrical double layer, suggested that the inhibitor molecules function by adsorption at the metal/solution interface. Thus, the change in C_{dl} values was caused by the gradual replacement of water molecules by the adsorption of the organic molecules on the metal surface, decreasing the extent of metal dissolution. In general, two modes of adsorption can be considered. The proceedings of physical adsorption require the presence of electrically charged metal surface and the charged species in the bulk of the solution. Chemisorption process involves charge sharing or charge transfer from the inhibitor molecules to the metal surface. This is possible in case of positive as well as negative charges on this surface. The presence of a transition metal, having vacant, low energy electron orbital, and an inhibitor molecule having relatively loosely bound electrons or hetero atoms with lone-pair electrons facilitates this adsorption [29, 30]. On the other hand, the inhibitor, which possesses nitrogen, sulphur and oxygen atoms with electron pair donors, can accept a proton, leading the cationic forms. These species can be adsorbed by the metal surface because of attractive forces between the negatively charged metal and the positively charged inhibitor molecules.

3.6. Inhibition mechanism

The mechanism of the inhibition process of corrosion of carbon steel in 1.0 M HCl solution by Schiff base compounds is mainly by adsorption one. The adsorption process is governed by different parameters that most depend on the chemical structure of these compounds. The inhibition efficiency of the additive compounds depends on many factors [31] which include the number of adsorption active centers in the molecule and their charge density, complex formation, molecular size and mode of interaction with metal surface. It is generally believed that the adsorption of the inhibitor at the metal/solution is the first step in the mechanism of inhibitors action in aggressive acidic solution.

The order of decreasing inhibition efficiency obtained from different techniques used in this study:

compound III > compound II > compound I

It is obvious from the above sequence that,compound III is more efficient inhibitors due to the presence of two OH group which is more electron donating. The electron donating groups enhance adsorption and increase the surface area covered by inhibitor. Compound II comes after compound III in the order of inhibition efficiency due to the presence of one OH group only. The compound I has the lowest inhibition efficiency due to the presence of Br ion which is an electron withdrawing group.

4. Conclusions

1. Schiff base compounds act as inhibitors for corrosion of 1018 carbon steel in 1 M HCl solution.
2. The percentage inhibition efficiency increased with increasing the concentration of the inhibitors and with decreasing temperature.
3. The inhibition process was explained in view of adsorption of Schiff base compounds on the steel surface.
4. The adsorption process obeyed Freundlich adsorption isotherm.
5. Polarization measurements showed that the inhibitors acted as mixed type inhibitors.
6. EIS measurements indicate that the single charge transfer process controlling the corrosion of carbon steel.

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