

PHYSICOCHEMICAL PROBLEMS
OF MATERIALS PROTECTION

Synthesis of Some Aromatic Nitro Compounds and its Applications
as Inhibitors for Corrosion of Carbon Steel
in Hydrochloric Acid Solution¹

M. Abdallah^{a, b}, B. H. Asghar^a, I. Zaafarany^a, and M. Sobhi^b

^a Department of Chemistry, Faculty of Applied Sciences, Umm Al-Qura University, Makkah, Saudi Arabia

^b Department of Chemistry, Faculty of Science, Benha University, Benha, Egypt

e-mail: metwally555@yahoo.com

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Abstract—The inhibition effect of some synthetic aromatic nitro compounds on the corrosion of carbon steel in 1 M HCl solution was studied using galvanostatic and potentiodynamic anodic polarization measurements. The percentage inhibition efficiency was found to increase with increasing the concentration of inhibitors and decreasing of temperature. Polarization data indicated that the additives acted as mixed-type inhibitors meaning that these compounds reduced the anodic dissolution of carbon steel and retard the cathodic hydrogen evolution reaction. Inhibition was interpreted in view of formation of insoluble complex adsorbed on the metal surface. The formation of complex was confirmed by UV-spectra. The adsorption of these compounds was found to obey Langmuir adsorption isotherm. Activation energy and some activated thermodynamic parameters were computed and discussed. It was found that these additives provide good protection to carbon steel against pitting corrosion in chloride-containing solution.

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1. INTRODUCTION

Corrosion of carbon steel is a fundamental academic and industrial concern that has received considerable amount of attention [1]. However, most equipment in industries is usually corroded owing to the general aggression of acid solutions. Some of the important fields of application of acid solutions in industries being acid pickling of iron and steel, chemical cleaning, ore production and oil well acidification. Thus, the use of inhibitors is one of the most practical methods for protection against corrosion in acidic media [2]. Different organic compounds has been studied as inhibitors to protect the steel from corrosive attack [3–12]. The inhibitive power of the organic compounds has been interpreted in terms of many different characteristics such as molecular size, molecular weight, molecular structure, nature of heteroatom present in the molecule, etc. [13]. Nitrogen containing compounds have been found to serve as good corrosion inhibitors and their inhibiting action has been explained in term of the number of mobile electron pairs [14] the π orbital character of free electron and the electron density around the nitrogen atom [15].

The aim of the present investigation is to examine the inhibitory action of some synthetic aromatic nitro compounds as inhibitors for the corrosion of carbon steel in 1 M HCl solution using galvanostatic polariza-

tion and electrochemical impedance spectroscopy techniques. The effect of temperature on the dissolution of carbon steel in free and inhibited acid solution was also studied and some thermodynamic parameters were calculated and discussed.

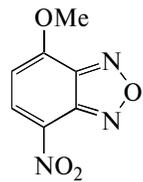
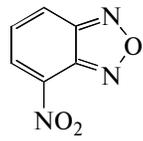
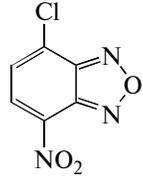
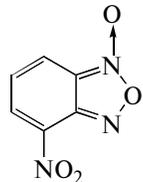
2. EXPERIMENTAL METHODS

The carbon steel specimen (L-52) used for this study has the following composition (wt %); C = 0.26; Mn = 1.35, P = 0.04, S = 0.05, Nb = 0.005, V = 0.02, Ti = 0.03, and Fe to balance. The galvanostatic polarization measurements were performed using specimens in the form of rods of 1 cm² exposed surface area as a working electrode. Electrical contacts were made through thick copper wires soldered to the end of electrodes not exposed to the solution. The electrode was successively abraded with the finest grade emery paper, degreased with acetone and finally washed with twice-distilled water, complete wetting of the surface was taken as indication of its cleanliness. All chemicals used were of A.R. quality. The solutions were prepared using twice-distilled water and no trials were made to deareate them. The electrolytic cell was all Pyrex and described elsewhere [16].

Galvanostatic polarization measurements were carried out using PS remote potentiostat with PS6 software for calculation of some corrosion parameters e.g., corrosion current density (I_{corr}), corrosion potential (E_{corr}) and Tafel constant (ba and bc). The corrosion parame-

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Table 1. Structure of the four synthesized aromatic nitro compounds

Compounds	Structure	Name
I		7-Methoxy-4-nitrobenzofurazan
II		4-Nitrobenzofurazan
III		7-Chloro-4-nitrobenzofurazan
IV		4-Nitrobenzofuroxan

ters were calculated from the intercept of the anodic and cathodic Tafel lines. A three compartment cell with a saturated calomel reference electrode (*SCE*) and a platinum foil auxiliary electrode was used. The inhibition efficiency *IE* was calculated using the equation:

$$\%IE = \left[\frac{I_{\text{free}} - I_{\text{add}}}{I_{\text{free}}} \right] \times 100, \quad (1)$$

where, I_{free} and I_{add} are the corrosion current densities in free and inhibited acid, respectively. Using UV-visible spectrophotometric method, some experiments were carried out on the electrolyte solution of the inhibited system before and after polarization measurements.

2.1. Synthesis of Inhibitors

Four compounds of aromatic nitro compounds were prepared and listed in Table 1.

Compound I: 7-Methoxy-4-Nitrobenzofurazan was prepared as follow, the solution of 7-chloro-4-nitrobenzofurazan (1.12 g, 5.622×10^{-3} mol) in methanol (50 cm^3) was added to 1mol dm^{-3} sodium methoxide solution (5.62 cm^3 , 5.62×10^{-3} mol). The mixture was heated at 40°C for 1 hour and then cooled and added to

iced water when 7-methoxy-4-nitrobenzofurazan precipitated; m.p. = 113°C , 115°C [17].

Compound II; 4-nitrobenzofurazan was prepared by nitration of benzofurazan using a 4 : 1 sulfuric:nitric acid mixture at 25°C . The resulting pale yellow precipitate was filtered under vacuum to give 4-nitrobenzofurazan in 63% yield, (mp $91-93^\circ\text{C}$, 93°C) [18].

Compound III: 7-Chloro-4-nitrobenzofurazan was a commercial sample.

Compound IV: 4-Nitrobenzofuroxan was prepared, as previously described [18], by nitration of benzofuroxan; mp: 143°C (mp 143°C) [19].

3. RESULTS AND DISCUSSION

3.1. Galvanostatic Polarization Measurements

Figure 1 shows the anodic and cathodic galvanostatic polarization curves of carbon steel electrode in 1 M HCl devoid of and containing different concentrations of compound I; for example. Similar curves were obtained for the other three compounds (not shown). Inspection of Fig. 1 shows that the presence of increasing concentration of the additive shifts the anodic curve to more positive potentials and the cathodic curve to more negative potentials. The corrosion current density (I_{corr}) was taken as the current of the point of intersection of the linear parts of cathodic and anodic curves. The potential of this intersection point is the corrosion potential (E_{corr}).

The corrosion parameters of carbon steel electrode in 1 M HCl solution containing different concentrations of inhibitors used were calculated and are presented in Table 2. Inspection of Table 2 reveals that, as the inhibitor concentrations increase, it is clear that, the anodic (*a*) and cathodic (*c*) Tafel slopes increase. This indicates that these compounds are mixed type inhibitors. However; the cathode was more polarized than the anode, meaning that the addition of these compounds to 1 M HCl solution reduced the anodic dissolution of carbon steel and retard the cathodic hydrogen evolution reaction. The values of E_{corr} are shifted slightly toward positive direction, $I_{\text{corr,decrease}}$ and hence the percentage inhibition efficiency ($\%IE$) increases. This indicates that the inhibitive effect of this compounds. This suggests that increase in inhibitor concentration increase the number of molecules adsorbed over the steel surface, blocking the active site of acid attack and thereby protecting the steel from corrosion. The order of the percentage inhibition efficiency decrease in the following sequence: Compound I > II > III > IV. This order will be discussed later.

3.2. Effect of Temperature

The effect of temperature on the anodic and cathodic galvanostatic polarization curves of carbon steel in 1 M HCl in the absence and presence of 500 ppm of inhibitors was studied at different temperatures

(30 to 60°C). Similar curves to Fig. 1 were obtained (not shown) and the corrosion parameters are given in Table 3.

Inspection of Table 3, it is clear that: as the temperature increases, the corrosion potential (E_{corr}) is shifted to more negative direction, the values of the corrosion current density (I_{corr}) increases and hence, the values of inhibition efficiency (IE) decreases. This indicates that the rising of temperature decreases the inhibition process due to the desorption of some adsorbed inhibitor molecules from the steel surface. This behavior proves that the adsorption of inhibitors on the carbon steel surface occurs through physical adsorption.

Activation parameters for corrosion of carbon steel were calculated from Arrhenius-type plot.

$$R_{\text{corr}} = A \exp(-E_a^*/RT) \quad (2)$$

and transition state- type equation:

$$R_{\text{corr}} = RT/Nh \exp(\Delta S^*/R) \exp(-H^*/RT), \quad (3)$$

where, R_{corr} is the rate metal dissolution and is directly related to corrosion current density I_{corr} [20], A is the frequency factor, N is Avogadro's number and R is the universal gas constant.

Figure 2 represents plot of $\log R_{\text{corr}}$ against $1/T$ for carbon steel in 1 M HCl solution in absence and presence of 500 ppm of inhibitors. Straight lines were obtained with slope equal to $-E_a^*/2.303R$. The values

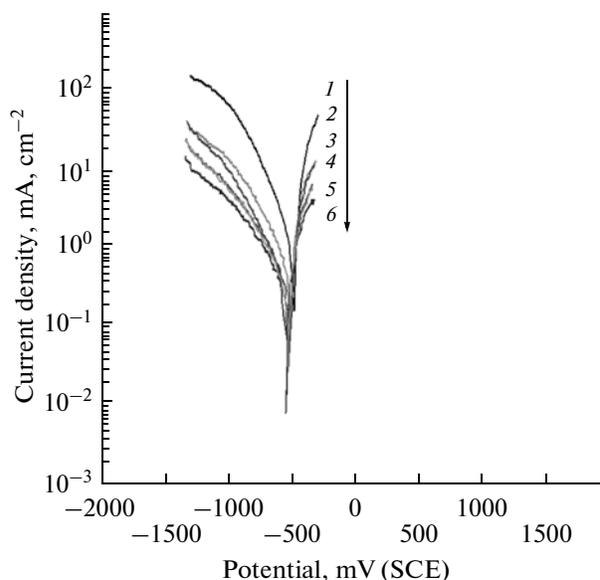


Fig. 1. Galvanostatic polarization curves of carbon steel in 1 M HCl containing different concentrations of compound I at 30°C. (1) 0.00 (2) 100 (3) 200 (4) 300 (5) 400 (6) 500 ppm.

of activation energy were found to be 22.48 kJ mol⁻¹ for 1 M HCl and equal to 28.82, 34.46, 38.29 and 42.12 kJ mol⁻¹ for compounds I, II, III and IV, respectively. The presence of aromatic nitro compounds

Table 2. Corrosion parameter obtained from galvanstatic polarization measurements of carbon steel in 1 M HCl solution containing different concentrations of inhibitors at 30°C

Comp.	Conc., ppm	$-E_{\text{corr}}$ mV (SCE)	I_{corr} mA cm ⁻²	β_c mVdec ⁻¹	β_a mVdec ⁻¹	% IE	0
Blank	—	495	1.32	288	77	—	—
I	100	566	0.47	292	82	64.39	0.64
	200	557	0.36	312	98	72.72	0.73
	300	551	0.22	342	103	83.33	0.83
	400	546	0.18	356	118	86.36	0.86
	500	549	0.11	369	125	91.66	0.92
II	100	558	0.52	312	88	60.60	0.61
	200	552	0.46	332	111	65.15	0.65
	300	546	0.28	345	125	78.78	0.79
	400	539	0.21	352	145	84.09	0.84
	500	534	0.16	360	168	87.87	0.88
III	100	555	0.62	310	95	53.03	0.53
	200	552	0.54	318	112	59.09	0.59
	300	546	0.38	328	128	71.21	0.71
	400	544	0.29	336	136	78.03	0.78
	500	547	0.22	348	156	83.33	0.83
IV	100	567	0.68	316	89	48.48	0.49
	200	561	0.59	332	122	55.30	0.55
	300	556	0.48	338	142	63.63	0.64
	400	548	0.34	345	155	74.24	0.74
	500	542	0.28	356	168	78.78	0.79

Table 3. Effect of temperatures on the corrosion parameters of carbon steel in 1 M HCl and 1 M HCl + 500 ppm of inhibitors

Temperature °C.	$-E_{\text{corr}}$ mV (SCE)	I_{corr} mA cm ⁻²	% IE
1M HCl			
30	495	1.32	—
40	496	1.48	
50	512	1.72	
60	528	1.98	
1M HCl + 500 ppm of inh.I			
30	549	0.11	91.66
40	562	0.18	84.74
50	572	0.25	76.41
60	578	0.32	65.22
1M HCl + 500 ppm of inh.II			
30	534	0.16	87.87
40	538	0.22	81.35
50	548	0.31	70.75
60	562	0.36	60.86
1M HCl + 500ppm of inh.III			
30	547	0.22	83.33
40	552	0.28	76.27
50	558	0.34	67.92
60	564	0.42	54.34
1M HCl + 500 ppm of inh.IV			
30	542	0.28	78.78
40	548	0.34	71.18
50	558	0.39	63.21
60	568	0.45	51.09

increases the activation energies of carbon steel indicating the adsorption of the inhibitor molecules on the metal surface and the presence of these additives induces energy barrier for the corrosion reaction.

On other hand, Fig. 3 represents of $\log R_{\text{corr}}/T$ against $1/T$ for carbon steel in 1 M HCl solution in absence and presence of 500 ppm of inhibitors. A straight lines relationship were obtained with slope equal $[-\Delta H^*/2.303 RT]$ and intercept equal $[\log(RT/Nh) - (\Delta S^*/20303R)]$. The values of ΔH^* obtained from the slope of straight equal to 23.65 kJ mol⁻¹ for 1M HCl and equal to 29.42, 36.22, 40.36 and 43.68 kJ mol⁻¹ or compounds I, II, III and IV, respectively. The values of H^* are different for studied compounds which mean that their structure affect the strength of its adsorption on the metal surface. The positive sign of enthalpy reflects the endothermic nature of the steel dissolution process [21]. The values of ΔS° calculated from the intercept of the straight line were found to be $-235.4 \text{ JK}^{-1} \text{ mol}^{-1}$ in 1 M HCl and equal to -252.2 , -248.6 , -236.2 and $-232.8 \text{ JK}^{-1} \text{ mol}^{-1}$ for compounds I, II, III, and IV respectively. The negative

values of ΔS^* in the absence and presence of the inhibitors implies that the activated complex in the rate determining step represent association rather than dissociation step, This means that the activated molecule were in higher order state than that at the initial stage [22].

3.3. Adsorption Isotherm

The efficiency of aromatic nitro compounds as a successful inhibitors is mainly dependent on its ability to get adsorbed on the metal surface. The adsorption of inhibitors molecules from an aqueous solution can be regarded as a quasi-substitution process between the organic compound in the aqueous phase, $\text{Org}(\text{aq})$, and water molecules at the steel surface, $\text{H}_2\text{O}(\text{sol})$ [23]



where, x the size ratio, is the number of water molecules displayed by one molecule of organic inhibitor.

The degree of surface coverage (θ) was evaluated at different concentrations of these additives in 1 M HCl solution using the following equation:

$$\theta = 1 - \frac{I_{\text{add}}}{I_{\text{free}}}, \quad (5)$$

where, I_{add} and I_{free} are defined previously. The values of θ have been inserted into Table 2. The degree of surface coverage θ was found to increase with increasing the concentration of aromatic nitro compounds. This indicates that the inhibitive action of these compounds toward the acid corrosion of carbon steel could be attributed to the adsorption of its compound onto the surface of the metal. The adsorbed layer acts as a barrier between the metal surface and aggressive solution leading to a decrease in the corrosion rate. Attempts were made to fit θ values to the several adsorption isotherms like Frumkin, Freundlich, Temkin and Langmuir. The best fit was obtained with Langmuir isotherm according to the following equation:

$$C/\theta = 1/K + C, \quad (6)$$

where, K is the equilibrium constant of adsorption.

The plotting of C/θ against C give straight line with unit slope, Fig. 4. This indicates that the adsorption of aromatic nitro compounds toward the steel surface in 1 M HCl solution follows Langmuir adsorption isotherm and consequently, there is no interaction between the molecules adsorbed at the metal surface. The values of equilibrium constant of adsorption (K) is calculated from the intercept and equal to 2.6×10^{-2} , 2.1×10^{-2} , 1.4×10^{-2} and 1.1×10^{-2} for compounds I, II, III, and IV respectively. The value of K is related to the standard free energy of adsorption. (G_{ads}) [24]

$$\ln K = \ln 1/55.5 - \Delta G_{\text{ads}}^\circ / RT, \quad (7)$$

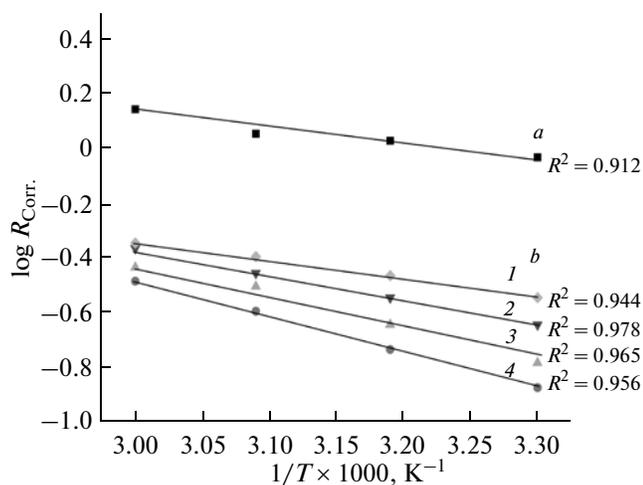


Fig. 2. Arrhenius plot ($\text{Log } R_{\text{Corr}}$ and $1/T$). For carbon steel in 1 M HCl in absence and presence of 500 ppm of inhibitor (a) 1 M HCl (b) 1 M HCl + 500 ppm of inhibitor, (1) Compound I (2) compound II (3) compound III (4) compound IV.

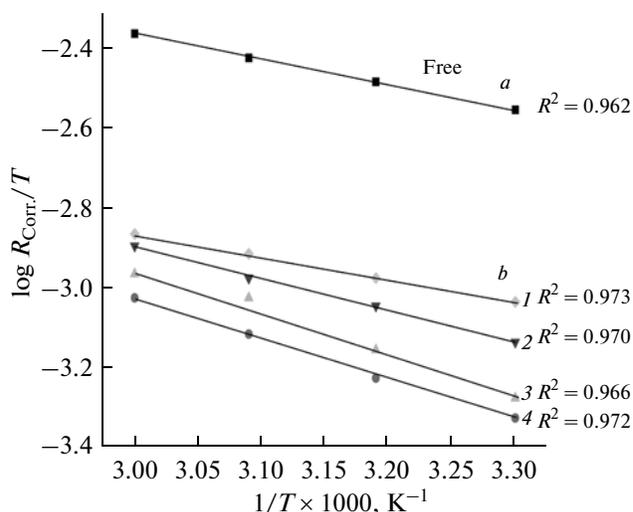


Fig. 3. Relation between $\text{Log } R_{\text{Corr}}$ and $1/T$ of carbon steel electrode in (a) 1 M HCl (b) 1 M HCl + 500 ppm of inhibitor (1) compound I (2) compound II (3) compound III (4) compound VI.

where, R is the universal gas constant (J mol^{-1}). The value 55.5 is the concentration of water in the solution in mole/l.

The calculated values of (G_{ads}) for aromatic nitro compounds adsorbed on the steel surface is -19.522 , -20.513 , -15.434 and -21.313 kJ mol^{-1} for compounds I, II, III, and IV respectively. It is of interest to mention here that the value of G_{ads} calculated in this work is not the exact value since the used concentration of inhibitors used in ppm not mol l^{-1} . However, the calculated value is enough to predict the type of adsorption process. The standard free energy change of adsorption is associated with water adsorption/desorption equilibrium which forms an important part in the overall free changes of adsorption. The negative value of G_{ads} obtained indicates that the physical adsorption process of this compound on the metal surface is spontaneous one.

3.4. Potentiodynamic Anodic Polarization Measurements

Figure 5 shows the potentiodynamic anodic polarization curves of carbon steel electrode in 1 M HCl + 0.05 M NaCl in absence and presence of different concentrations of compound I as an example of the tested compounds at a scanning rate 1 mV/s. NaCl Similar curves were obtained for other compounds (not shown). NaCl was used as pitting corrosion agent. The potential was swept from negative potential towards anodic direction up to the pitting potential. No any anodic oxidation peaks are observed in all anodic scan. The pitting potential (E_{pitt}) was taken as the potential at which the current owing, along the passive film increases suddenly to higher values, denoting the destruction of passive film and initiation of visible pits. It was found that increasing the concentrations of these compounds cause a shift of

the pitting potential in the noble direction indicating an increase of the resistance to pitting attack.

The effect of addition of increasing concentrations of aromatic nitro compounds on the values of pitting potential is illustrated in Fig. 6. This figure represents the relationship between E_{pitt} and logarithm of molar concentration of additives. It is clear from this figure that, as the concentration of additives increases, the pitting potential shifted to more positive values in accordance with the following equation:

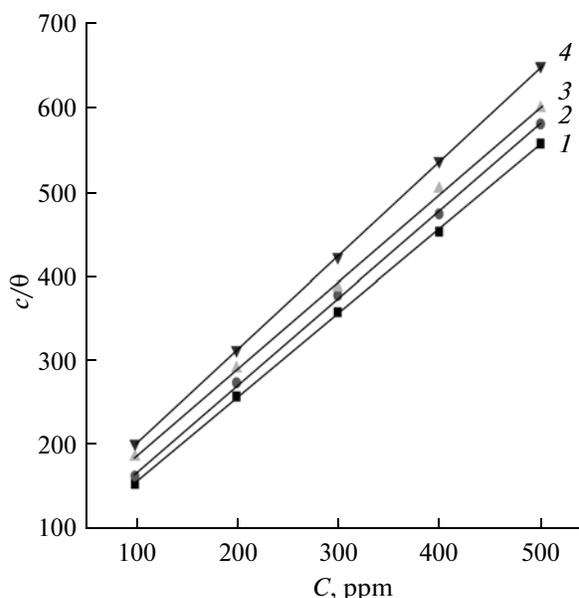


Fig. 4. Langmuir adsorption isotherm plotted as $\text{Log } C/\theta$ and $\text{log } C$. (1) compound IV (2) compound III (3) compound II (4) compound I.

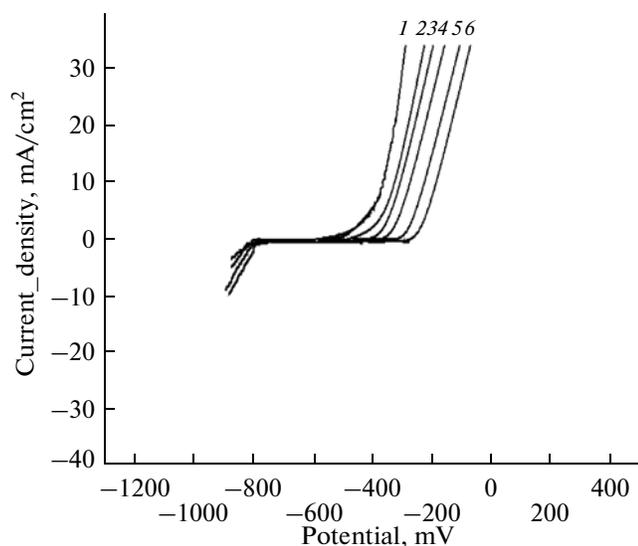


Fig. 5. Potentiodynamic anodic polarization curves of carbon steel in 1 M HCl solution + 0.5 M NaCl containing different concentrations of compound I at a scan rate 1 mV sec^{-1} . (1) 0.00 (2) 100 (3) 200 (4) 300 (5) 400 (6) 500 ppm.

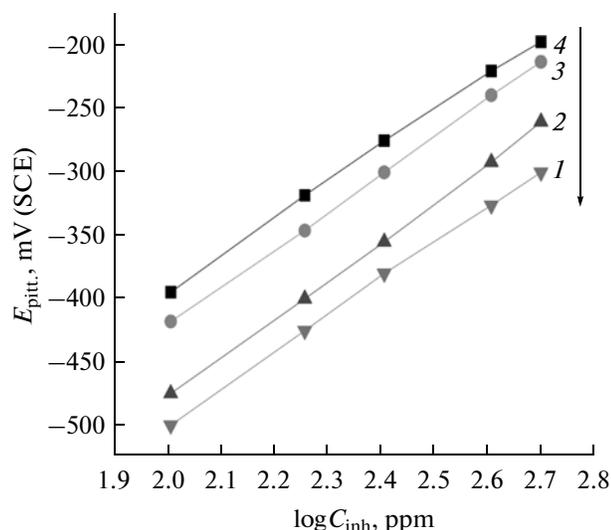


Fig. 6. The relation between pitting potential of C-steel and logarithm the concentration of inhibitors (1) compound IV (2) compound III (3) compound II (4) compound I.

$$E_{\text{pitt}} = a + b \log C_{\text{inh}}, \quad (8)$$

where, a and b are constants depending on the type of additives used and the nature of the electrode. The positive shift of E_{pitt} indicates the increase resistance to pitting attack. At one and the same inhibitor concentration the marked shift of potential in the positive direction (increased resistance to pitting corrosion) decreases in the following sequence:

compound I > compound II > compound III > compound IV

There is a good agreement in the order of inhibition efficiency between two different techniques used.

3.5. Mechanism of Inhibition

Corrosion inhibition of carbon steel in hydrochloric acid solution by aromatic nitro compounds can be explained on the basis of molecular adsorption. These compounds inhibit the corrosion by controlling both anodic as well as cathodic reactions. The process of adsorption is governed by different parameters almost depend on the chemical structure of these inhibitors. The high performance of all compounds is attributed to the presence of π electrons, large molecular size, presence of nitrogen and oxygen atoms in the chemicals structure of inhibitors and the ability to form complexes [25]. The inhibition mechanism of these compounds under investigation is due to the formation of complex between ferrous ion and these compounds. The formed complex is adsorbed on the metal surface and thereby isolating the metal from further corroding attack. To provide an evidence for formation of complex, the UV-visible spectrophotometric measurements was used. Inspection of the chemical structure of all represented compounds, the molecules have structures characterized by the presence of chelating center represented by oxygen and nitrogen atoms as extra sources of lone pair of electrons. The formation of complexes of these compounds with ferrous ions released during the corrosion reaction is also considered. In order to confirm the possibility of formation of compound-Fe complex, UV-visible absorption spectra obtained from the corrosive solution. Figure 7 represents the UV-spectra of the compound I in 1 M HCl solutions after measurements in: (1) compound I (2) Fe^{+2} cation, (3) Fe^{+2} cation + compound I. Similar curves were obtained for other three compounds used (not shown).

The electronic absorption spectra of these compounds display two bands in UV-visible region. The shorter wavelength band with λ_{max} at 205 nm is ascribed to $\pi - \pi^*$ transition of the benzenoid system of the compound. The second band (curve 1) with λ_{max} at 333 nm can be attributed to $\pi - \pi^*$ transition within the heterocyclic moiety of the compound I. The localization of this band at longer wavelength, relative to the former one, can be ascribed to the higher delocalization of π —electrons of heterocyclic moiety. The band appears (curve 2) at λ_{max} 675 nm is due to ferrous ion whereas the band with λ_{max} 660 nm may be due to the formation of metal complexes between ferrous ion and compound I (curve 3), it is clearly seen that the band maximum of $\pi - \pi^*$ transition within the heterocyclic moiety underwent a blue shift, suggesting the interaction between compound I and Fe^{+2} ions in the solution. Compounds II, III and IV cause a shift in wavelength and the increase in absorbance gives a strong evidence of complex formation between these compounds and Fe^{+2} ions. As the strength of metal complex increases the blue shift increase.

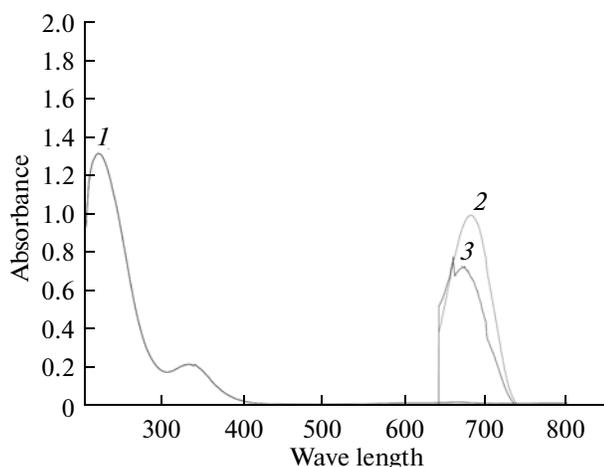


Fig. 7. UV-spectra of the additives used as inhibitors for carbon steel corrosion in 1 M HCl solutions after measurements: (1) compound I, (2) Fe^{+2} cation, (3) Fe^{+2} cation + compound I.

The mode of adsorption depends on the affinity of the metal toward the electron clouds of the ring system. The order of inhibition efficiency of the investigated compounds decrease in the following order: Compound I > II > III > IV. Compound I exhibits higher inhibition efficiency due to the presence of electro donating group $-\text{OCH}_3$ group (+I, +M) with negative Hammett constant ($\sigma = -0.27$), so the group will increase the electron charge density on the molecule which facilitate the adsorption process. Compound II comes after compound I and has no substituent (H atom with $\sigma = 0.0$), which contributes no effect on the charge density of the molecule. Compound III comes after compound I and II due to the presence of $-\text{Cl}$ ($-I, +M$) electro withdrawing group and has positive Hammett constant ($\sigma = +0.23$) i.e., a group that lowers the electron density on the molecule and hence lowers the inhibition efficiency. Compound IV is the lowest inhibition efficiency due to the presence of N^+ as ammonium group ($-I, -M$) which decrease the electron density on the ring.

4. CONCLUSIONS

1—The investigated aromatic nitro compounds act as an inhibitors for corrosion of carbon steel in 1 M HCl.

2—The inhibition efficiency increased with the increase of inhibitor concentration but decreases with an increase in the temperature.

3—The compounds are of mixed-type inhibitors.

4—The inhibitive action of these compound takes place through the adsorption of their molecules on the carbon steel surface.

5—The adsorption of these compounds obeys Langmuir isotherm.

6—Aromatic nitro compounds protect the pitting corrosion of carbon steel in chloride containing solution.

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