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Fluorenone Hydrazone Derivatives as efficient Inhibitors of Acidic and Pitting Corrosion of Carbon Steel

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Abstract

The inhibiting effect of some fluorenone hydrazone derivatives toward the corrosion of carbon steel in 1.0 MH_2SO_4 solution was studied using galvanostatic polarization and potentiodynamic anodic polarization techniques. The inhibition percentage efficiency increases with increasing the concentration of the studied inhibitors but decreases with rising temperature and the presence of the electron withdrawing groups. Polarization curves revealed that the fluorenone hydrazone derivatives act as mixed-type inhibitors. The inhibition process was interpreted in view of its adsorption at the carbon steel surface. The adsorption process obeyed Freundlich adsorption isotherm. Fluorenone hydrazone derivatives increase the resistance to pitting assault in chloride solutions by shifting the pitting corrosion potential to more positive directions.

Keywords: Fluorenone Hydrazone Derivatives, corrosion inhibitors, pitting corrosion, Adsorption

1. Introduction

Fluorene and its derivatives (FLs) are a unique class of are polycyclic aromatic hydrocarbons (PAHs) exist in the fossil fuels, petrogenic sources and among products from the burning of gasoline [1]. Fluorene hydrazone derivatives of carbonyl compounds considered to be biologically important class of compounds [2]. Hydrazone nucleus is found in natural and synthetic products of biological interest [3]. Literature studies revealed that, various substituted hydrazones are associated with a broad spectrum of biological activities [4-6]. Carbon steel Type (L-52) has found large applications in various industries process especially in the manufacture of Egyptian petroleum pipes lines. Sulfuric acid is generally used for elimination of objectionable scale and rust in several industrial processes especially in the pickling processes of steel. Inhibitors are one of the most practical methods used to prevent steel dissolution and acid consumption [7]. Several organic molecules are used to inhibit the corrosion attack of carbon steel in acidic solutions [8-27]. Most of organic molecules inhibit the steel dissolution by adsorption on the metal surface and blocking the active corrosion sites. Adsorption of the organic compounds on the steel surface represents the most important action of these compounds. The strength of adsorption depends on the physico-chemical properties of the organic molecules such as function group aromaticity, electron density of the donor atom, the electronic structure of the molecules and π-orbital character of the donating electrons [28]. Some organic and natural compounds are used to inhibit the pitting corrosion of some metals and alloys by shifting the pitting corrosion potential to more positive (noble) direction [29-34].This manuscript will shed more light on other application of some fluorenone hydrazones derivatives as efficient corrosion inhibitors for carbon steel in 1.0M H_2SO_4 solution using galvanostatic and potentiodynamic anodic polarization measurements. Hopefully, this will allow the development of new strategy for the development of novel compounds toward materials and corrosion inhibitors. To follow the multiaddressable applications of hydrazone derivatives and in continuation of work on the uses of fluorenone hydrazones as precursors for the synthesis of photochromic di and tetrahydroindolizines [5,6], this manuscript will shed more light on other application of some fluorenone hydrazone derivatives as efficient corrosion inhibitors for carbon steel in 1.0M H_2SO_4 solution using galvanostatic and potentiodynamic anodic polarization measurements. This will permit the growth of innovative new strategies for the development of novel compounds toward materials and corrosion inhibitors.

2. Experimental techniques

Carbon steel of type (L-52)used in this study having the chemical composition (0.26% C, 1.35% Mn,0.04%, P , 0.03% Ti, 0.02% V, 0.05% S,0.005% Nb and the remainder Fe). For galvanostatic polarization measurements, the electrodes were polished with different grades emery papers (grade 400,800, 1200 and 1500), degreased with acetone and rinsed with double distilled water. For electrochemical cell, three electrodes was used .The working electrode is a cylindrical rod of carbon steel embedded in araldite with exposed surface area 0.56cm^2 used as working electrode. A saturated calomel reference electrode (SCE) and platinum foil auxiliary electrode were used as the reference and counter electrode, respectively. Galvanostatic and potentiodynamic anodic polarization measurements were carried out using a PS remote potentiostat with PS6 software, for calculation the corrosion current density (I_{corr}), corrosion potential (E_{corr}), anodic and cathodic Tafel constants (b_a and b_c). The corrosion parameters were calculated from the intercept of the anodic and cathodic Tafel lines. The potential was measured against a saturated calomel electrode (SCE) as reference. All measured solutions were freshly prepared using analytical grade chemicals and distilled water.

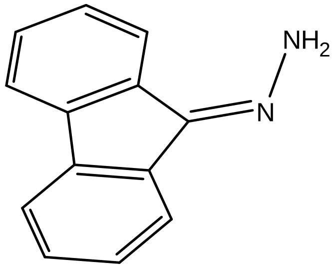
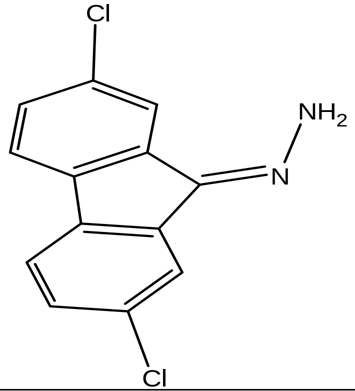
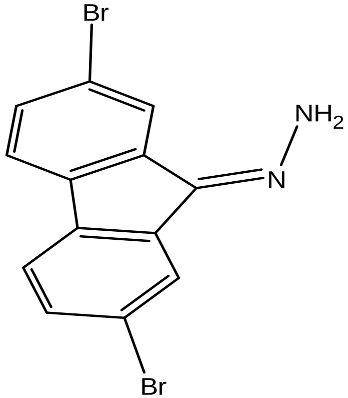
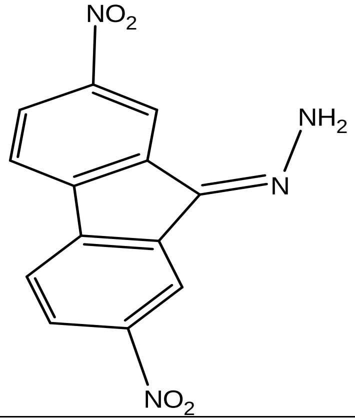
2.1. *Inhibitors*

9H-Fluorenone was purchased from Aldrich. 2,7-dichloro-9H-fluoren-9-one ,2,7-dibromo-9H-fluoren-9-one and 2,7-dinitro-9H-fluoren-9-one were prepared according the literature procedures [35-40] with some modifications through the reaction with NCS, NBS and HNO_3/H_2SO_4, respectively. Condensation of the fluorenone derivatives with Hydrazine hydrate in refluxing ethanol and catalytic amount of acetic acid for 8 h, afforded the desired hydrazones (Table 1) in moderate to good yield (53-84%). The pure products were obtained after recrystallization from ethanol in case of hydrazones I and II and in acetic acid in case of hydrazone III and IV. All solvents used were of spectroscopic grade and used without further purifications.

2.2. Instrumentation

The synthesized fluorenone hydrazones were confirmed by both spectroscopic and analytical tools. All melting points were measured on a Gallenkamp melting point apparatus. The IR spectra were monitored in potassium bromide discs on FT-IR spectra were recorded on Shimadzu IRAfinity-1S infrared spectrophotometers using KBr pellet technique. NMR was recorded on a Bruker Advance 400 MHz with $CDCl_3$ in case of hydrazone and DMSO in case of hydrazones as solvents with tetramethylsilane (TMS) as the internal reference.

Table1: Chemical structures, names, molecular weights, molecular formulas of the fluorene hydrazone derivatives **I-IV**

Compound no.	**Structure**	**Chemical name**	**Molecular formula**	**Molecular Weight**
I		(9H-fluoren-9-ylidene)hydrazine	$C_{13}H_{10}N_2$	194.23
II		(2,7-dichloro-9H-fluoren-9-ylidene)hydrazine	$C_{13}H_8Cl_2N_2$	263.12
III		2,7-dibromo-9H-fluoren-9-ylidene)hydrazine	$C_{13}H_8Br_2N_2$	349.91
IV		(2,7-dinitro-9H-fluoren-9-ylidene)hydrazine	$C_{13}H_8N_4O_4$	284.23

3. Results and discussion

3.1. Galvanostatic polarization

Fig.1. shows the galvanostatic polarization curves for C-steel electrode in 1.0 M H_2SO_4 solution in the presence and absence of different concentrations of compound I as an example of the studied inhibitors .Similar curves were recorded for the three other compounds (not shown).The corrosion parameters , e.g. corrosion current density (I_{corr}), corrosion potential (E_{corr}), anodic and cathodic Tafel constants (b_a and $b_{c'}$,percentage inhibition efficiency (%I.E) and surface coverage (θ) were calculated from the galvanostatic polarization curves and given in table 2. The percentage inhibition efficiency (% IE) and a parameter (θ) which represents the part of the metal surface roofed by the inhibitor molecules were calculated using the following equations:

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

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

where, $I_{corr\text{-}free}$ and $I_{corr.add}$ are the corrosion current densities in both free and inhibited solution, respectively.

An inspection of the results obtained from figure 1 and Table 2 it is clear that, as the concentration of the of fluorenone hydrazone derivatives increases, the anodic and cathodic curves exhibit Tafel-type behavior . Tafel lines are shifted to more negative and more positive potentials relative to the blank curve. The values of anodic and cathodic Tafel slopes (β_a & β_c) are increases. This indicated that these compounds act as mixed type inhibitor mainly cathodic because the cathode is more polarized when an external current was applied ($\beta_c > \beta_a$).The values of $E_{corr.}$ are approximately constant, the values of I_{corr} decreases and the values of percentage inhibition efficiency increases. This indicates that, the inhibiting effect of fluorenone hydrazone derivatives [41]. At one and the same inhibitor concentration, the inhibition percentage efficacy decreases in the following sequence:

compound I > compound II > compound III > compound VI . This sequence will be discussed later.

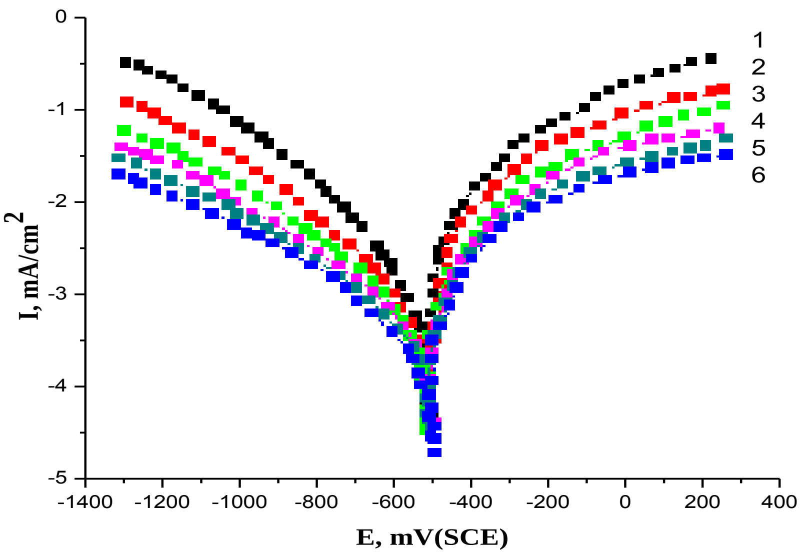


Fig. 1. Galvanostatic polarization curves of carbon steel in 1.0 M H_2SO_4 containing different concentrations of compound I (1) 0.00 (2)200 (3) 400 (4)600 (5) 800 (6)1000 ppm

Table 2. Corrosion parameters obtained from galvanostatic polarization curves of carbon steel in 1.0M H_2SO_4 and containing dissimilar concentrations of the inhibitors

Concn. **ppm**	**$-E_{corr}$** **mV(SCE)**	**I_{corr}** **m A cm^{-2}**	**β_a** **m V dec^{-1}**	**β_c** **m V dec^{-1}**	**% *IE***	**θ**
Compound I						
0.00 ppm	504	1.22	68	318	-	-
200 ppm	505	0.38	82	326	68.85	0.689
400 ppm	508	0.31	98	344	74.59	0.746
600 ppm	508	0.22	126	362	81.96	0. 820
800 ppm	510	0.09	148	388	92.62	0.926
1000ppm	510	0.06	172	420	95.08	0.951
Compound II						
200 ppm	504	0.42	78	328	65.57	0.656
400 ppm	505	0.36	96	336	70.49	0.705
600 ppm	507	0.24	113	350	80.33	0.803
800 ppm	509	0.12	130	402	90.16	0.902
1000ppm	508	0.08	162	424	93.44	0.934
Compound III						
200 ppm	505	0.46	80	316	62.29	0.623
400 ppm	506	0.42	96	322	65.57	0.656
600 ppm	508	0.27	124	344	77.86	0.779
800 ppm	508	0.20	138	364	83.61	0.836
1000ppm	506	0.13	162	386	89.34	0.893
Compound VI						
200 ppm	503	0.54	88	328	55.73	0.557
400 ppm	506	0.48	98	343	60.65	0.607
600 ppm	508	0.35	122	365	71.31	0.711
800 ppm	507	0.26	136	372	78.69	0.787
1000ppm	508	0.19	156	380	84.43	0.844

3.2. Effect of Temperature

The effect of rising temperature on the corrosion of C-steel in 1.0M H_2SO_4 solution devoid of and containing different concentrations of the fluorenone hydrazine derivatives over the temperature range from 30 to 60 ^oC was studied using galvanostatic polarization measurements. Similar curves to figure 1 were obtained (not shown). The corrosion parameters e.g .E_{corr}, I_{corr} and % IE was calculated and given in Table. It is clear that from table 3, the values of $E_{corr.}$is shifted slightly to more negative values. The values of $I_{corr.}$ increase and hence the values of % IE decreases indicating the desorpotin of these compounds at high temperature.

The activation energy (E^*_a) of the corrosion process was calculated using Arrhenius equation:

$$R_{corr.}=A \exp (-E_a^*/ RT) \qquad (3)$$

where R_{corr} is the rate of corrosion and is directly related to corrosion current density I_{corr} [42], A is the Arrhenius constant, R is the gas constant and T is the absolute temperature.

Figure 2 shows the relation between the log of $R_{corr.}$ and1/T(Arrhenius plot) in free and inhibited 1.0 M H_2SO_4 containing 1000 ppm of the studied fluorenone hydrazone derivatives. Straight lines were obtained. The values of E_a can be calculated from the slope of the straight lines and equal to 41.68 kJ mol^{-1} in 1.0 M H_2SO_4 and equal to 47.42, 49.93, 53.24 and 55.97 kJ mol^{-1} in presence of compounds, I , II, III, and IV, respectively. The values of E^*_a in presence of the fluorenone hydrazone additives are higher than obtained in free solution suggesting that dissolution of C-steel is sluggish in the attendance of inhibitor due to physical adsorption [43]. This can be attributed to the formation of a film on the C-steel surface helping as an energy barrier for the C-steel corrosion. The enthalpy (ΔH^*) and entropy (ΔS^*) of activation of the corrosion process were calculated from the following equation:

$$R_{corr} = (RT/ Nh) \exp (\Delta S^*/R) \exp (-\Delta H^*/RT) \qquad (4)$$

where, h is Planck's constant and N is Avogadro's number.

A plot of log (R_{corr}/ T) vs 1/ T for C-steel in free and inhibited 1.0 MH_2SO_4 containing 1000 ppm of the studied organic compounds, gives straight lines as shown in Figure 3.

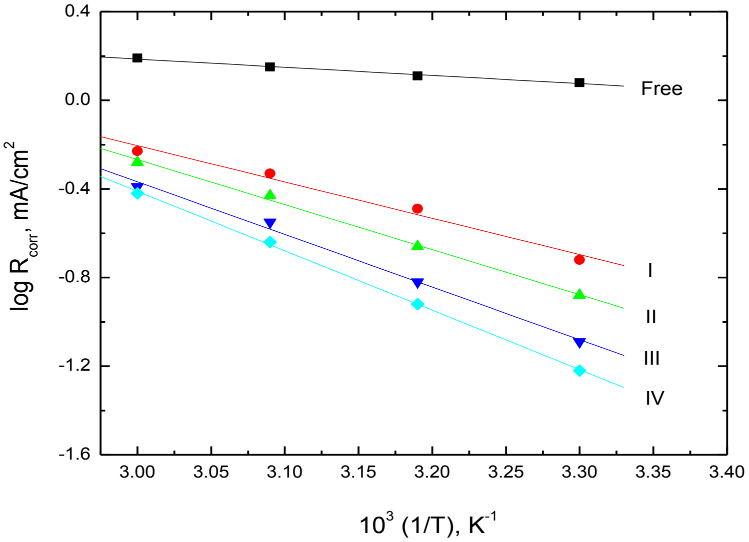


Fig.2: Relation between log R_{corr} and the reciprocal of temperature of carbon steel electrode in a) 1.0M H_2SO_4; b) 1.0M H_2SO_4 +1000 ppm of the studied compounds

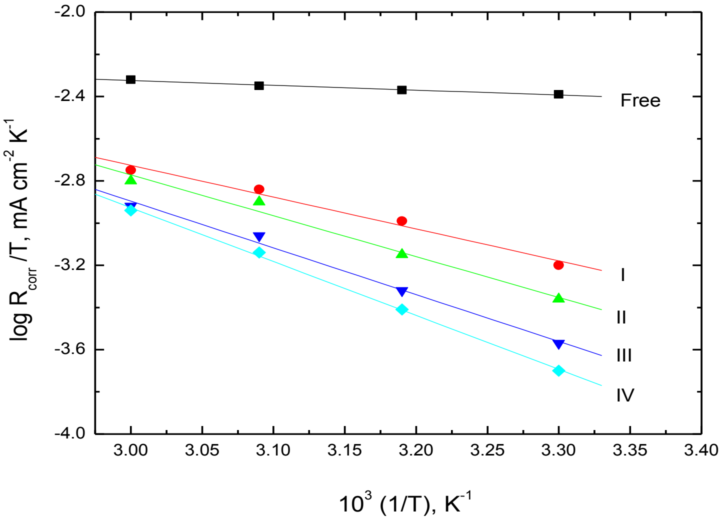


Fig.3: Relation between log R_{corr}/T and the reciprocal of temperature of carbon steel electrode in a) 1.0 M H_2SO_4 b) 1.0 M H_2SO_4 +1000 ppm of the studied compounds.

Table 3. Effect of temperatures on the corrosion parameters of carbon steel in 1.0 M H_2SO_4 and 1.0M H_2SO_4+1000ppm of inhibitors

T°C Temperature ^oC.	$-E_{corr}$ mV (SCE)	I_{corr} mA cm^{-2}	% IE
1.0M H_2SO_4			
30°C	504	1.22	-
40°C	508	1.28	-
50°C	509	1.42	-
60°C	510	1.58	-
1.0M H_2SO_4+1000ppm of of inh.I			
30°C	508	0.06	95.08
40°C	510	0.12	90.62
50°C	512	0.23	83.80
60°C	513	0.38	75.94
1.0M H_2SO_4+1000ppm of inh.II			
30°C	506	0.08	93.44
40°C	509	0.15	88.28
50°C	511	0.28	80.28
60°C	512	0.40	74.68
1.0M H_2SO_4+1000ppm of inh.III			
30°C	508	0.13	89.34
40°C	509	0.28	78.12
50°C	512	0.42	70.42
60°C	514	0.52	67.08
1.0M H_2SO_4+1000ppm of inh.IV			
30°C	510	0.19	84.42
40°C	508	0.32	75.00
50°C	512	0.46	67.60
60°C	514	0.58	63.29

The values of ΔH^* obtained from the slope of the straight line and equal to 38.45kJ mol^{-1} in1.0 M H_2SO_4 and equal to 43.33,45.97,47.56,50.78 and 52.22 kJ mol^{-1} in presence of compounds, I ,II,III and IV respectively. The measured positive signs of ΔH^* reflect the endothermic nature of the steel dissolution process. The values of ΔH^* are different for the compounds under investigation which mean that, their structure affect the strength of its adsorption on the metal surface. The values of ΔS^* calculated from the intercept of the straight line and equal to 164.56 J mol^{-1} K$^{\mathbf{-1}}$in1.0M H_2SO_4 and equal to 186.56,198.92, 212.64 and 223.84J mol^{-1} K$^{\mathbf{-1}}$ in presence of compounds, I , II, III and IV, respectively.

The negative values of ΔS^* in the presence and absence of the inhibitors implies that, the activated complex is the rate determining step and reflects association rather than dissociation. It's also reveals that, the noticeable increase in the reaction order takes place from reactants to the activated complex.

3.3. Inhibition of pitting corrosion

Fluorenone hydrazones derivatives were tested as pitting corrosion inhibitors using potentiodynamic anodic polarization techniques.Fig.4 shows the effect of increasing concentration of compound I as an example of the tested compounds on the potentiodynamic anodic polarization curves of carbon steel in 1.0 M H_2SO_4 containing 0.5 M NaCl solution as a pitting corrosion agent at scan rate 1 mV/sec. The same figures were obtained for the other studied compounds not shown. It is obvious that, there is no any anodic dissolution peak observed during the anodic scan due to the thickness and the hardness of passive film. At certain potential the current increases rapidly to higher values due to the breaking the passive film and the occurrence of pitting corrosion. This potential is known pitting corrosion potential (E_{pitt}.) As the concentration of fluorenone hydrazones derivatives increases, the pitting corrosion potential shifted to more positive direction .This proves that the studied compounds inhibit the pitting corrosion of carbon steel in chloride containing solutions. The inhibition of the pitting corrosion by the fluorenone hydrazones derivatives can be explained as competitive adsorption between the fluorenone hydrazones derivatives and Cl^- ions. If the fluorenone hydrazones derivatives adsorption is predominate on the surface. This cause the inhibition of pitting corrosion .On the other hand, if the adsorption of Cl^- ions predominates, pitting corrosion will occur on the surface.

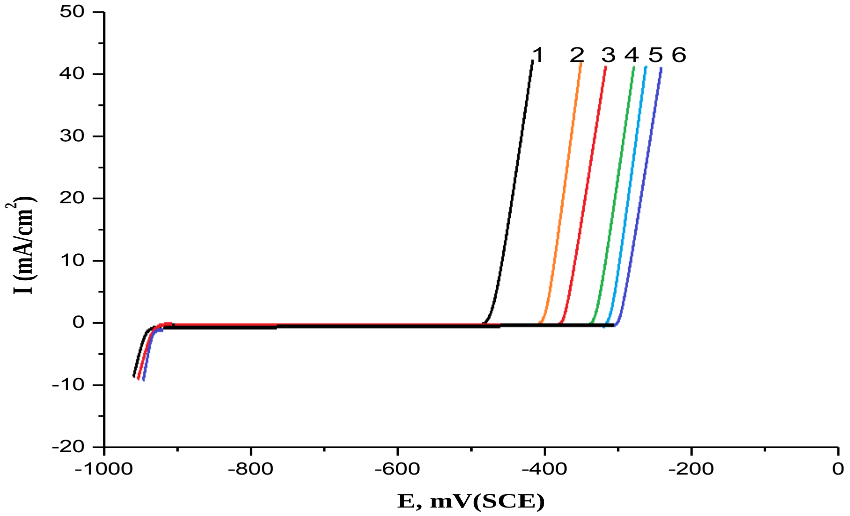


Fig.4. Potetiodynamic anodic polarization curves of carbon steel in 1.0 M H_2SO_4 solution + 0.5M NaCl containing different concentrations of compound I at a scan rate1mVsec^{-1}, (1) 0.00 (2)200 (3) 400 (4)600 (5) 800 (6)1000 ppm.

The relation between the pitting corrosion potential and the logarithmic concentrations of the studied inhibitors was illustrated in Fig.5. It is clear that from Fig 6, as the concentration of the inhibitors increases the $E_{pitt.,}$ is shifted to more positive values satisfying the following equation[44,45]:

$$E_{pitt.} = a + b \log C_{inh.} \quad (5)$$

where a and b are constants depending on the types of electrode and inhibitors used. At one and the same inhibitors concentration, the more positive shift in the noble direction (more resistance to pitting corrosion) decrease in the following sequence:

compound I > compound II > compound III > compound VI

The sequence of the % IE of the fluorenone hydrazine derivatives obtained from galvanostatic polarization is in a good agreement with the positive shift in the values of $E_{pitt.}$ obtained from potentiodynamic anodic polarization measurements.

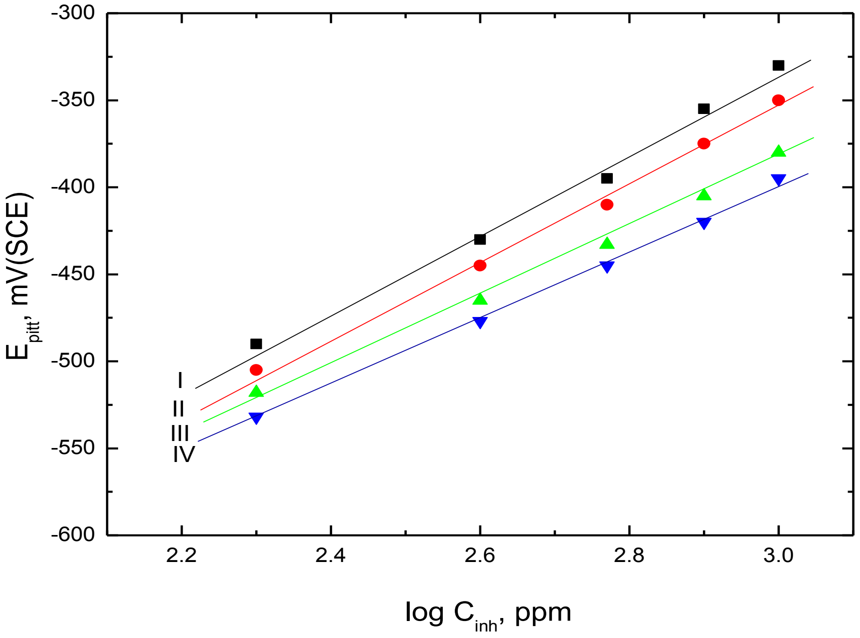


Fig.5. The relation between pitting potential of carbon steel and the logarithmic of the concentration of inhibitors

3.4.Mechanism of inhibition and adsorption isotherm

The efficiency of fluorenone hydrazone derivatives to inhibit the corrosion of carbon steel in sulfuric acid solution is mainly dependent on its ability to form adherent adsorbed layer on the metal surface

The adsorption can be regarded as a quasi-substitution process between the fluorenone hydrazine derivatives in the aqueous phase[$Fh_{(sol)}$] and water molecules at the electrode surface [$H_2O_{(ads)}$] according to the following equation.

$$Fh_{(sol)} + n\,H_2O_{(ads)} \text{ -------------- } Fh_{(ads)} + n\,H_2O_{(sol)} \qquad (3)$$

where, n is the size ratio, that is ,the number of water molecules replaced by one organic inhibitor. Fh is the fluorenone hydrazone derivatives adsorbed at the steel surface. The interaction between the inhibitor and the steel surface can be examined by the adsorption isotherm .The values of surface coverage, θ, for different concentration of the studied fluorenone hydrazone derivatives have been used to explain the best isotherm to determine the adsorption process.

In order to obtain a suitable adsorption isotherm, attempts were made to fit θ to various isotherms e.g., Temkin, Frumkin, Langmuir, Freundlich, Flory-Huggins and Bockris-Swinkels, By far the best results of investigated inhibitors were fitted by Freundlich adsorption isotherm equation [46-48] which is given as:

$$\theta = K\,C^n \qquad (4)$$

or alternatively, by

$$\log \theta = \log k + n \log C \qquad (5)$$

where k and C represent the equilibrium constant of adsorption process and inhibitor concentration, respectively.

Fig. 6 represents the relation between the log θ and log C. Straight lines with intercept of log k were obtained indicating that the adsorption of these compounds on the steel surface follows Freundlich adsorption isotherm. The calculated values for the equilibrium constant of adsorption were found to be (0.25, 1.85, 3.98, 10.19) x10^{-3}, for compounds I, II, III and IV, respectively.

The standard free energy of adsorption $\Delta G°_{ads}$ can be calculated from the value of the equilibrium constant using the following equation [49]:

$$K= 1/55.5 \exp [-\Delta G°_{ads}/RT] \qquad (6)$$

where, 55.5 is the concentration of water in solution in mol/l, R is the universal gas constant and T is the absolute temperature. The values of $\Delta G°_{ads}$ equal to -1.4,-2.5,-4.5,-5.8 and -7.6 kJmol^{-1}, for compounds I, II, III and IV, respectively. The negative values of $\Delta G°_{ads}$ indicate the spontaneous adsorption of the inhibitor on the metal surface and also the interaction between the studied fluorenone hydrazine derivatives and the steel surface is strong

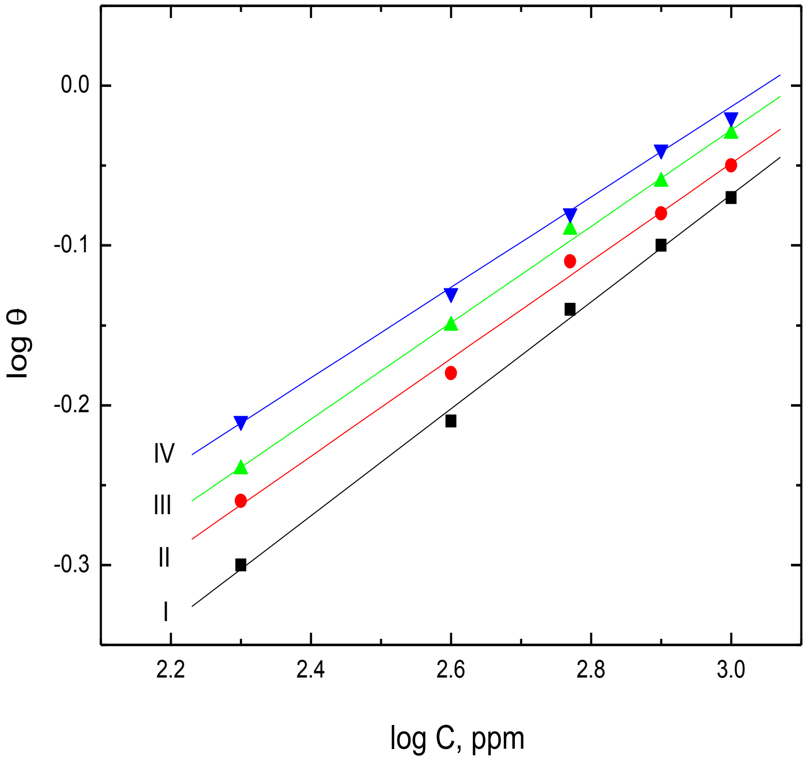


Fig.6. The relation between logarithm of surface coverage (θ) and logarithm of inhibitor concentration (C). (Freundlich adsorption isotherm)

From the results obtained from galvanostatic polarization and potentiodynamic anodic polarization techniques. The sequence of the percentage inhibition efficiency decreases in the following order:

Compound I > Compound II > Compound III > Compound VI

Compound IV showed the lowest inhibition efficiency due to the presence of nitro group as electron attracting property. The presence of the nitro group decreases the adsorption of the compound on the steel surface which led to the decrease of the electron deficiency of the bonding between the compound and the iron surface. On the other hand, chloro and bromo substituted floureneone hydrazones as in compounds II and III found to be more efficient compared with compound IV. Interestingly, chlorinated fluorenone hydrazone (compound II) was found to be more efficient than bromo substituted fluorenone hydrazone (compound III) irrespective to the high electronegativity of chlorine atom. This may be attributed to the bigger size of the bromine atom (steric effect). In addition, the highest inhibition efficiency is observed in the case on unsubstituted fluorenone hydrazone (compound I) which found to be 95.08% at 1000ppm.

Conclusions

The fluorenone hydrazone derivatives were found to be effective inhibitors of corrosion of carbon steel in 1.0 MH_2SO_4 solution as investigated using galvanostatic polarization and potentiodynamic anodic polarization techniques. A directly proportional between the percentage inhibition efficiency and the concentration of the studied inhibitor but inversely proportional with rising temperature and the presence of the electron withdrawing groups were monitored. Polarization curves revealed that the fluorenone hydrazone derivatives act as mixed-type inhibitors. The inhibition process was interpreted in view of its adsorption at the carbon steel surface. In addition, the adsorption process obeyed Freundlich adsorption isotherm. Finally, fluorenone hydrazone derivatives increase the resistance to pitting attack in chloride solutions by shifting the pitting corrosion potential to more positive directions.

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