

Corrosion inhibition of Isosorbidedinitrate drug on Carbon steel in Hydrochloric Acid solution

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Abstract

The inhibitory effect of isosorbidedinitrate drug (ISDN) on carbon steel was demonstrated in 1M HCl solution by weight loss, potentiodynamic polarization and electrochemical impedance measurements. The inhibition of ISDN drug is increased with increased concentration and minimize temperature. Data obtained from the polarization mention that ISDN drug is a mixed type inhibitor. The inhibition power of ISDN drug was explained on the basis of its adsorption on the carbon steel surface and the corrosion of carbon steel is lowered by adding ISDN drug. The adsorption of ISDN drug on to the surface of carbon steel in 1 M HCl obeys the Langmuir isotherm. Also, activation thermodynamic parameters such as E_a , ΔH^* and ΔS^* were calculated using Arrhenius and transition state equations and discussed.

Keywords: Isosorbidedinitrate, Corrosion inhibition, EIS, Adsorption.

1. Introduction

Corrosion of carbon steel has been widely studied due to its industrial importance, in different acidic, alkaline, and salt as aggressive corrosion media [1]. The corrosion of metallic surfaces in acidic solutions causes considerable costs. In order to reduce the corrosion of metals, several techniques have been applied. The use of corrosion inhibitors is one of the most practical methods for protection against corrosion in acidic media [2,3]. Corrosion can be defined as the destructive attack of a material (mostly metal) by reaction with its environment [4]. In last few years, scientists interested in finding an efficient technique for treatment and prevention of metal from corrosion one of these techniques is corrosion inhibitors which offer surface protection by adsorption of their active functionalities on metal surfaces [5-7].

Currently in recent years, many scientists have tended to use some pharmaceutical compounds to be used as an inhibitor for corrosion of certain metals and alloys [8,11]. Such drugs are cheap, safe, and easily adsorbed due to the presence of active centers for the adsorption. In previous research, an antibiotic such as gentamicin, kanamycin and amikacin, antihypertensive, antibacterial,azole antifungal drugs were utilized as inhibitor for the dissolution of Al in acid solutions [12,14].

In this work, the isosorbidedinitrate drug (ISDN) was investigated as corrosion inhibitors for carbon steel in 1 M HCl. The inhibitory action of Isosorbidedinitrate (ISDN) was investigated by various techniques (electrochemical impedance spectroscopy (EIS), potentiodynamic polarization and the weight loss.

2. Experimental

2.1. Carbon steel

Carbon steel specimens with chemical composition (wt %) were used in the experiments: Carbon (C) 0.38-0.45, Chromium (Cr) 0.90-1.20, Molybdenum (Mo) 0.15-0.25, Silicon (Si) 0.17-0.37, Manganese (Mn) 0.50-0.80 and balance Fe.

A pre-treatment procedure was carried out, prior to each experiment, in which the surface of specimen of (1.6 x 1.5 x 0.7) cm was mechanically polished with different emery paper and cleaned with acetone and distilled water then dried at room temperature before use.

2.2 Weight loss technique

Carbon steel specimens were washed, dried, and accurately weighed and then the specimens were immersed in a solution containing 1 M HCl with and without the addition of different concentrations of inhibitor (50:250 ppm) for 24 h. After immersion time, the specimens were washed, dried, and weighed accurately.

2.3 Electrochemical technique

Electrochemical measurements were measured using (Metrohm Autolab PGSTAT 204). A classical three electrode glass cell with a platinum counter electrode and saturated calomel electrode (SCE) as a reference was used. Carbon steel as working electrode expose only a 0.48 cm² surface to the solution. The exposure surface was abraded with different grades of emery paper, washed with acetone and distilled water then dried. The potentiodynamic polarization measurements were obtained using scan rate of 2 mV s⁻¹ at 25 ± 1 °C. The electrochemical impedance spectroscopy (EIS) measurements were carried out as described elsewhere. The degree of surface coverage (θ) and the inhibition efficiency (IE %) were calculated.

2.3 Inhibitor

Isosorbidedinitrate (ISDN) is a medication used for heart failure, esophageal spasms, and to treat and prevent chest pain from not enough blood flow to the heart. The chemical Formula of isosorbidedinitrate is C₆H₈N₂O₈. Fig1. Shows the Structure of isosorbidedinitrate (ISDN) which contain hetero atoms (N and O), double bonds (N=O) and positive charge on N-atoms besides negative charge on O-atom.

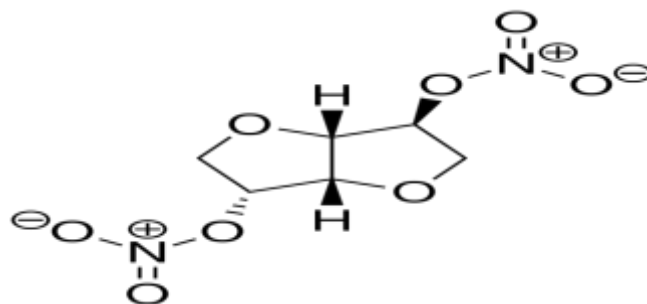


Fig (1) Structure of isosorbidedinitrate (ISDN)

3.Results and Discussion

3.1 Weight loss measurements

The change in weight was recorded. The weight loss (ΔW , in grams) is given by equation [15]:

$$\Delta W = (W_1 - W_2)$$

Where, W_1 and W_2 are the weight of specimen before and after the reaction, respectively. The corrosion rate (r , in $\text{mg cm}^{-2} \text{d}^{-1}$) was calculated according to the following equation [16]:

$$r = \Delta W / St$$

Where ΔW is the weight loss of carbon steel sheets, S is the total area of the specimen, and (t) is immersion time. The inhibition efficiencies ($IE\%$) of inhibitor were determined from equation [15]:

$$IE\% = [(\Delta W_{\text{free}} - \Delta W_{\text{inh}}) / \Delta W_{\text{free}}] \times 100$$

Where, ΔW_{inh} and ΔW_{free} are the weights loss of specimen in presence and absence of different

concentrations of inhibitor (50:250 ppm), respectively. The weight loss of carbon steel, corrosion rate values and the inhibition efficiencies ($IE\%$) were calculated and listed in Table (1). The data given in Table (1) shows that the corrosion rate values decreased as the concentrations of the inhibitor is increased.

Fig (2) represents the relation between inhibition efficiencies and concentrations at different times. The figures show that, the corrosion inhibition was increased by increasing the inhibitor concentration. This behavior was due to the adsorption and the coverage of inhibitor on the carbon steel surface. This adsorption decreases the dissolution of carbon steel surface by blocking the corrosion sites and hence decreasing the weight loss while increasing the efficiency as concentration of inhibitor increases [17].

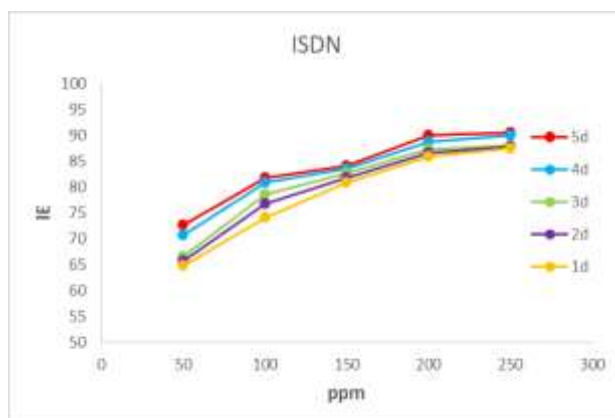


Fig (2) relation between inhibition efficiencies and concentrations at different times of ISDN.

Table (1) values of corrosion rate and inhibition efficiency in absence and presence of different concentrations of ISDN in 1 M HCl solution.

ISDN	1D			2D			3D			4D			5D		
Conc.	Δw	R	$IE\%$	Δw	r	$IE\%$	Δw	r	$IE\%$	Δw	r	$IE\%$	Δw	r	$IE\%$
Blank	0.0688	6.41	---	0.1007	4.69	----	0.1301	4.04	----	0.1556	3.62	---	0.1741	3.24	---
50	0.0243	2.26	64.68	0.0345	1.61	65.73	0.0436	1.35	66.48	0.0456	1.06	70.69	0.0476	0.88	72.65
100	0.0178	1.66	74.12	0.0234	1.09	76.76	0.0279	0.86	78.55	0.0299	0.69	80.78	0.0319	0.59	81.67
150	0.0132	1.23	80.81	0.0183	0.85	81.82	0.0227	0.70	82.55	0.0257	0.59	83.48	0.0277	0.51	84.08
200	0.0097	0.90	85.90	0.0135	0.62	86.59	0.0166	0.51	87.24	0.0176	0.41	88.68	0.0176	0.32	89.89
250	0.0086	0.80	87.50	0.0122	0.56	87.88	0.0156	0.48	88.01	0.0156	0.36	89.97	0.0166	0.31	90.46

3.2. Potentiodynamic polarization technique

The representative potentiodynamic polarization curve of carbon steel in 1 M HCl solution in the absence and presence of various concentrations of the inhibitor is shown in Fig (3). Some corrosion kinetics parameters, such as corrosion potential (E_{corr}), cathodic and anodic Tafel slopes (β_c & β_a), corrosion current density (I_{corr}) achieved from the extrapolation of the anodic and cathodic polarization curves, corrosion rate (r) and inhibition efficiency (IE %) were computed and presented in Table (2). The degree of surface coverage (Θ) and the inhibition efficiency (IE %) were calculated as following [18] :

$$\Theta = \frac{(I_{corr}^{free} - I_{corr}^{inh})}{I_{corr}^{free}}$$

$$IE \% = (\Theta) \times 100$$

Where, I_{corr}^{free} and I_{corr}^{inh} are the corrosion current densities for carbon steel electrode in absence and presence of the inhibitors. Data in Table (2) reveals that, when the concentration of the synthesized inhibitor was increased, the inhibition efficiencies increased while the corrosion current densities decreased. This increase in inhibition efficiency with increasing inhibitor's concentration indicated that the inhibitor was acting as an adsorption inhibitor. The inhibitive action was discussed in terms of blocking the electrode surface by adsorption of the molecules through the active centers contained in its structure as an electrostatic interaction between positive centers (N^+) in inhibitor and charged sites on the metallic surface and formation of protective (insoluble) layer on metal surface in presence of inhibitor. This protection layer on the metal surface, decreases the contact between the metal surface and the aggressive medium. This inhibitor causes change in the anodic and cathodic Tafel slopes and no definite trend

was observed in the shift of E_{corr} values in the presence of different concentrations of the inhibitor, suggesting that this inhibitor behaves as mixed-type inhibitor. The values of the cathodic Tafel slope (β_c) and the anodic Tafel slope (β_a) for the inhibitor were shifted slightly. The slight variations in the Tafel slope suggested that the inhibitor is blocking the cathodic and anodic sites without changing the corrosion mechanism [17].

3.3 Effect of temperature

The effect of temperature on the corrosion of carbon steel in 1 M HCl solution in absence and presence of high concentration of inhibitor was studied using potentiodynamic polarization technique.

Figs (4,5) represent the potentiodynamic polarization plots for carbon steel electrode in 1M HCl in the absence and presence of high concentration of inhibitor at scanning rate 2 mV/sec at different temperature. Some corrosion kinetics parameters, such as corrosion potential (E_{corr}), cathodic and anodic Tafel slopes (β_c & β_a), corrosion current density (I_{corr}) achieved from the extrapolation of the anodic and cathodic polarization curves, corrosion rate (r) and inhibition efficiency (IE %) were computed and presented in Tables (3,4) which reveal that:

- The values of corrosion rate increase with increase of temperature
- Increasing temperature has almost no effect on corrosion potential.

The corrosion efficiency decreases as temperature increases which indicate formation of adsorptive film of physical character [17].

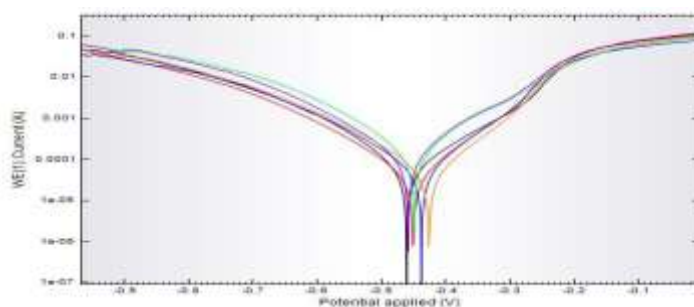


Fig (3) potentiodynamic polarization curves of carbon steel in 1 M HCl solution in absence and presence of different concentrations of ISDN

Table (2) corrosion parameters of carbon steel in 1 M HCl solution in absence and presence of different concentrations of ISDN.

ISDN	ba (V/dec)	bc (V/dec)	E_{corr} Calc (V)	j_{corr} (A/cm ²)	r (mm/year)	Θ	IE
Blank	0.13188	0.10402	-0.44494	0.00013307	1.5463	----	----
50	0.11006	0.084316	-0.42296	5.85E-05	0.67048	0.560	56.02
100	0.092911	0.082514	-0.46117	4.59E-05	0.53197	0.65	65.48
150	0.08003	0.078734	-0.45299	3.45E-05	0.39521	0.74	74.07
200	0.10662	0.085736	-0.43768	2.92E-05	0.33044	0.78	78.01
250	0.10929	0.085168	-0.44781	2.47E-05	0.289554	0.81	81.39

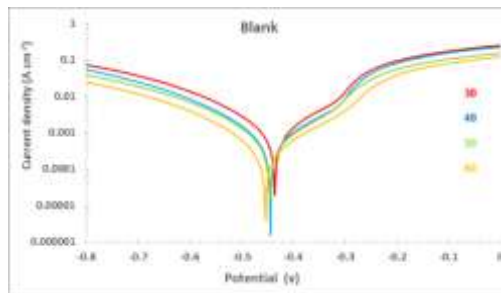


Fig (4) Potentiodynamic polarization curves of carbon steel in 1 M HCl solution at different temperatures in absence of ISDN.

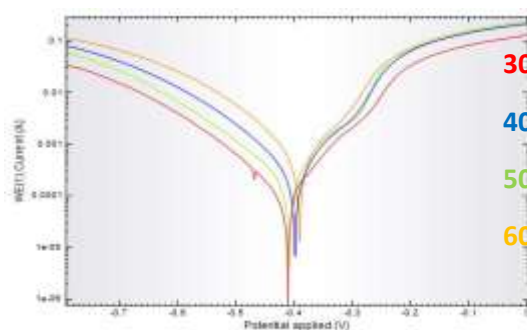


Fig (5) Potentiodynamic polarization curves of carbon steel in 1 M HCl solution in presence of ISDN at different temperatures.

Table (3) Corrosion parameters of carbon steel in 1 M HCl solution in at different temperatures in absence of ISDN.

Blank	β_a (V/dec)	β_c (V/dec)	E_{corr} (V)	j_{corr} (A/cm ²)	r (mm/year)
30	0.10	0.18	-0.41	0.000701	8.14
40	0.15	0.08	-0.41	0.000935	10.86
50	0.09	0.16	-0.41	0.001189	13.81
60	0.12	0.11	-0.43	0.001555	18.06

Table (4) corrosion parameters of carbon steel in 1 M HCl solution in presence of ISDN at different temperatures.

ISDN	b_a (V/dec)	b_c (V/dec)	E_{corr} (V)	j_{corr} (A/cm ²)	r (mm/year)	Θ	IE
30	0.097	0.072	-0.41	1.33E-04	1.56	0.809	80.98
40	0.082	0.072	-0.39	0.00019262	2.23	0.793	79.39
50	0.107	0.076	-0.41	0.00029586	3.44	0.751	75.11
60	0.103	0.078	-0.38	0.00045585	5.29	0.706	70.68

3.4 Kinetic parameter

The activation energy (E_a) for the corrosion of carbon steel in 1 M HCl solution in absence and presence of the inhibitor at different temperatures were calculated from Arrhenius Equation [17, 19]:

$$r = A e^{(-E_a/RT)}$$

And the logarithmic form:

$$\ln r = \ln A - (E_a/RT)$$

Where, r represents rate of corrosion reaction, A is the Arrhenius constant, R is the gas constant and T is the absolute temperature. Arrhenius plots of $\ln r$ vs. $1/T$ gave straight line, as shown graphically in Fig (6), with linear regression coefficients are very close to 1, indicating that

the corrosion of steel in 1 M HCl without and with inhibitor follows the Arrhenius equation with slope of ($-E_a/R$). Activation energies were calculated and listed in

Table (5). The data shows that the activation energy (E_a) of the corrosion of carbon steel in 1 M HCl solution in the presence of the inhibitor was higher than that in free acid solution, indicating that the inhibitor was adsorbed on the steel surface physically [17]. The change in enthalpy and entropy of activation values (ΔH^* , ΔS^*) were calculated from the transition state theory [20].

$$\ln (r/T) = [\ln(R / N_A h) + (\Delta S^* / R)] - (\Delta H^* / RT)$$

Where, h is the Plank constant, N_A is the Avogadro number, R is the ideal gas constant, ΔH^* is the enthalpy of activation and ΔS^* is the entropy of activation. Plotting of $\ln(r/T)$ versus $(1/T)$, gave straight lines as shown in Fig(7). with slope of $-(\Delta H^*/R)$ and the intercept of $\ln(R/N_A h) + (\Delta S^*/R)$. Values of ΔH^* and ΔS^* were calculated and listed Table (5).

The positive sign of the enthalpy of activation (ΔH^*), reflecting the endothermic nature of the corrosion process and means that the dissolution of carbon steel is difficult in the presence of inhibitor. The negative sign of entropy of activation (ΔS^*), indicates that the activated complex in the rate determining step represents an association rather than dissociation, reflecting that more order take place, going from reactant to activate complex [17].

3.5 Adsorption isotherm

The adsorption of the inhibitor was accompanied by desorption of water molecules from the surface. The degree of surface coverage (θ) for different concentrations of the inhibitor were evaluated from the potentiodynamic polarization data. The values of surface coverage (θ) of different concentrations of the inhibitor have been used to explain the isotherm for adsorption of the inhibitor on the carbon steel surface.

Several adsorption isotherms were tested to describe the adsorption behavior of the inhibitors used in this study. The Langmuir isotherm is the best description of adsorption behavior of the inhibitor molecules on carbon steel surface according to the following equation: [21]

$$C/\theta = (1/K_{ads}) + C$$

Where: C is the concentration of inhibitor, K_{ads} is the equilibrium constant of the adsorption process and (θ) is the surface coverage.

Fig (8) represents plots of C/θ versus C which gave a straight line with intercept of $(1/K_{ads})$, and the correlation coefficient (R^2) equal to 1. This indicates that, the adsorption of inhibitor on the carbon steel surface in 1M HCl solution follows Langmuir's adsorption isotherm [17].

The free energy of adsorption (ΔG^0_{ads}) was calculated from the following equation: [22]

$$\Delta G^0_{ads} = -RT \ln (55.5 K_{ads})$$

Where, the value (55.5) is the molar concentration of water in solution in molarity units (mol L^{-1}). The values of K_{ads} and ΔG^0_{ads} , were calculated and listed Table (6).

The value of ΔG^0_{ads} up to -20 kJ mol^{-1} or lower were consistent with the electrostatic interaction between charged organic molecules and the charged metal surface (physical adsorption); while those about -40 kJ mol^{-1} or higher were involved sharing or transferring a lone pair of electrons from the organic molecules to the metal surface to form a coordinate type of bond (chemisorption). The value of ΔG^0_{ads} in Table (6) indicates that the adsorption process of inhibitor on metal surfaces is physical adsorption. The negative sign of G^0_{ads} suggests that the adsorption of inhibitor onto the carbon steel surface is a spontaneous process and usually characteristic of strong interaction and a highly efficient adsorption [17, 23,24]

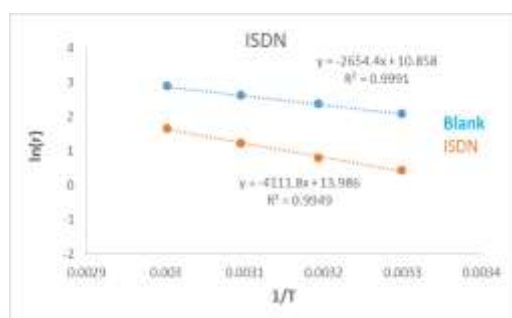


Fig (6) Arrhenius plots of carbon steel in 1 M HCl solution in absence and presence of ISDN at different temperatures.

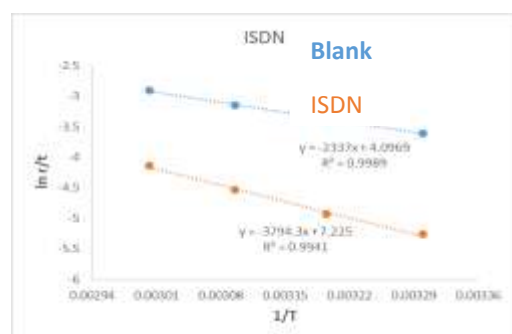
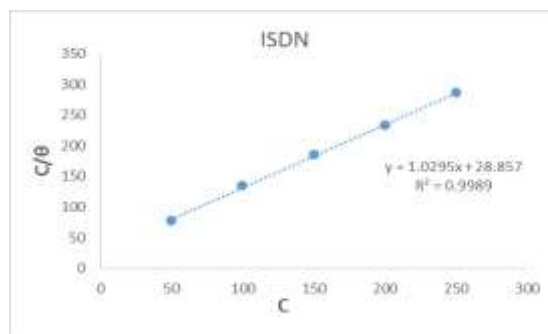


Fig (7) Transition state plots of carbon steel in 1 M HCl solution in absence and presence of ISDN at different temperatures.

Table (5) Activation thermodynamic parameters of carbon steel in 1 M HCl in absence and presence of ISDNat different temperatures.

	E_a (J/mol)	ΔH^* (J. mol)	ΔS^* (J. mol)
Blank	22068.89	19429.66	-163.458
ISDN	34185.11	31545.88	-137.452

**Fig (8)** Langmuir adsorption isotherm plots for the adsorption of ISDN in 1 M HCl on carbon steel surface.**Table (6)** adsorption of thermodynamic parameters of ISDN on carbon steel surface.

	Slope	intercept	k_{ads}	ΔG_{ads} (kJ/mol)
ISDN	1.029468	28.85661186	0.034654	-13.5

3.6. Electrochemical impedance spectroscopy (EIS) technique

The corrosion behavior of carbon steel in 1M HCl solution in the absence and presence of different concentrations of the inhibitor at 25 °C ± 1 was studied using EIS (Electrochemical impedance spectroscopy) technique. The degree of surface coverage (θ) and the inhibition efficiency (IE %) were calculated using charge transfer resistance as follow [15, 24] :

$$(\theta) = (R_{ct}^{inh} - R_{ct}^{free}) / R_{ct}^{inh}$$

$$IE \% = (\theta) \times 100$$

Where, R_{ct}^{free} and R_{ct}^{inh} are the charge transfer resistance values in the absence and presence of inhibitor for carbon steel in 1M HCl. The degree of surface coverage (θ), the inhibition efficiency (IE %) and charge transfer resistance values were listed in Table (7).

Fig (9), shows the Nyquist plots of carbon steel in 1M HCl solution in absence and presence of different concentrations of the inhibitor. The Nyquist impedance doesn't show perfect semicircle which attributed to the frequency dispersion as a result of roughness and inhomogenates of electrode surface. The impedance diagram shows the same trend (one capacitive loop), however, the diameter of this capacitive loop increases with increasing the concentration of inhibitors, giving consequently a decrease in the corrosion rate. The increase in R_{ct} value is due to the adsorption of inhibitor on the metal surface [25,26]. The inhibition efficiency (IE %) and the degree of surface coverage (θ) increase as concentration of inhibitors increases. This fact suggests that the inhibitor molecules may first be adsorbed on the steel surface and cover some sites of the electrode surface by formation of a protective layer [17].

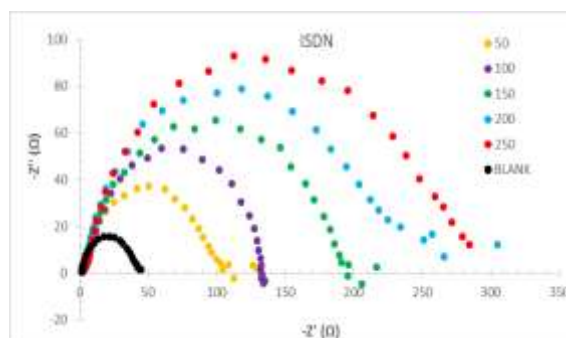
**Fig (9)** Nyquist diagrams of carbon steel in 1 M HCl solution in absence and presence of different concentrations of ISDN.

Table (7) Impedance of carbon steel in 1 M HCl solution in absence and presence of different concentrations of ISDN.

	$R_{ct} (\Omega)$	$R_s (\Omega)$	θ	IE
Blank	39.56	1.8138	---	---
50	99.551	2.4345	0.60	60.26
100	129.9	3.7325	0.69	69.54
150	194.87	0.7816	0.79	79.69
200	223.67	1.5434	0.8231	82.313
250	271.46	1.7194	0.8543	85.427

4. Conclusion

- The results obtained lead to the conclusion that ISDN drugs effectively inhibit the corrosion of carbon steel in 1 M HCL solutions.
- The adsorption model obeys the Langmuir isotherm. The negative values of ΔG_{ads} indicate that the adsorption of the inhibitor molecule is a spontaneous process and an adsorption mechanism follows physical (ionic) processes.
- Polarization curves demonstrated that the ISDN is a mixed type inhibitor for carbon steel surface.
- EIS plots indicated that the addition of inhibitors increases the charge-transfer resistance and shows that the inhibitive performance depends on adsorption of the molecules on the metal surface.
- The inhibition efficiencies determined by weight loss, potentiodynamic polarization and EIS techniques are in reasonably good agreement.

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