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Expired Indomethacin Therapeutics as Corrosion Inhibitors for Carbon Steel in 1.0 M Hydrochloric Acid Media

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Abstract

Indomethacin in the form of expired drugs which have low price was used as corrosion inhibitor for carbon steel in 1.0 M HCl solution using chemical and electrochemical techniques, namely, weight loss, electrochemical impedance spectroscopy (EIS), potentiodynamic polarization, and scanning electron microscopy (SEM). Inhibition efficiency increased with increasing inhibitor concentrations and decrease by temperature. The Results show that expired indocin capsules is a good inhibitor where the inhibition efficiency reaches 83% at 500 ppm of the used inhibitors. Data obtained from EIS studies were analyzed to model the corrosion inhibition process through appropriate equivalent circuit models. The adsorption of indocin drugs obeyed Langmuir adsorption isotherm. The thermodynamic parameters were calculated and discussed. Polarization curves indicated that they are mixed type of the inhibitors. SEM was used to study the surface morphology of the steel in the absence and presence of the additive. The results obtained from weight loss, EIS, and Potentiodynamic polarization are in good agreement and show that the expired drugs of many advantages like low price, save, and high availability act as eco-friendly corrosion inhibitor for steel

Keywords Corrosion · Carbon steel · Drugs · Weight loss · EIS · SEM

1 Introduction

Organic corrosion inhibitors were applied extensively to protect steel from corrosion in many aggressive acidic media (e.g., in the acid pickling and cleaning processes of metals) [1–12]**.** The effectiveness of these compounds as corrosion inhibitors has been interpreted in terms of their molecular structure, molecular size, and molecular mass, heteroatom present and adsorptive tendencies [13]. The existing data show that most organic inhibitors act by adsorption on the metal surface. Quite a number of studies have been carried out in determination of adsorptive of various compounds at the electrode/solution interface [14–16]**.** A large number of organic compounds were studied as corrosion inhibitor, unfortunately most of the organic inhibitors used are very expensive and health hazards. Their toxic properties limit their field of application. Thus, it remains an important object to find cost-effective and nonhazardous inhibitors for the protection of metals against corrosion. In this connection, the influences of nontoxic organic compounds and drugs on the corrosion of metals in acid media were investigated by several authors [17–25]. A.M. Fekry et al., have been studied the effect of natural products as eco-friendly corrosion inhibitors for mild steel in different corrosive medium by chemical and electrochemical measurements [26–29]**.** The environmentally safe materials, uracil and adenine, were tested experimentally as inhibitors for the corrosion of 70Sn–Ag alloy in aggressive nitric acid solution by A.M. Fekry and coworkers using electrochemical techniques, and Theoretical Study [29]**.** The use of expired drugs as corrosion inhibitors can be traced back to 2009s by R. S. Abdel Hameed, where expired ranitidine was used as corrosion inhibitors for Al in HCl corrosive medium [30, 31].

In 2011s, R.S. Abdel Hameed [32] reported the use of expired ranitidine drugs as nontoxic corrosion inhibitor for mild steel in hydrochloric acid medium. The use of the expired drugs as corrosion inhibitors is of high advantages,

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such as (1) it low price, (2) high availability, (3) save and eco-friendly, (4) nontoxic inhibitors, (5) decrease the solid waste accumulation by prevention of the expired drugs accumulation to the environment, and (6) better than the old traditional inhibitor which is difficult in their preparation and application. The objective of this work is to study the corrosion inhibitive action of expired indomethacin -SR drugs for the corrosion of carbon steel in 1.0 M HCl. The choice of this drug as a corrosion inhibitor is based on its environmentally friendly; its molecule has O, N atoms as active center atoms. The survey of literature reveals that indocin-SR capsules is belongs to a group of medicines called Nonsteroidal Anti-Inflammatory Drugs (or NSAID), it is used to relieve pain and reduce inflammation (swelling, redness, and soreness), Each indocin capsule contains 50 mg of indomethacin as the active ingredient. It also contains the inactive ingredients: Lactose, lecithin, silica-colloidal anhydrous, magnesium stearate, gelatin, titanium dioxide, iron oxide yellow CI77492, and opacode monogramming ink S-1-17823 black. So the uses of indocine capsules as corrosion inhibitor are promise.

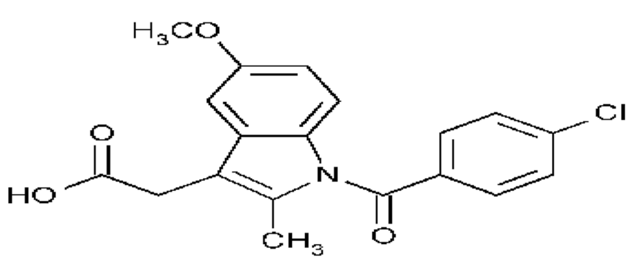
2 Experimental

2.1 Material

Analytical grade HCl and double distilled water were used to prepare all solutions. Indocin-SR of molecular formula: $C_{19}H_{16}ClNO_4$ and molecular weight: 357.8, from (EIPICO) pharmaceutical company, was used in the form of indocine capsules, for the study as in the following chemical structure.

2.2 Weight Loss Techniques

Carbon steel alloy is with the following chemical composition: wt% of 0.29 C, 1.25 Mn, 0.03 P, 0.03 Sn, 0.04 Cr, 0.04 Mo, and 0.027 Si and the remainder iron. The test coupons were cut into $2 \times 2 \times 0.2$ cm^3. The samples were first mechanically polished with a fine grade emery paper in order to obtain a smooth surface, followed by degreasing in acetone [33–36] then rinsed in distilled water and finally dried between two filter papers and weighed. All experimental were carried out at 25, 35, 45, and 55 ± 1 °C.



Chemical structure of the used inhibitor indocin-SR

2.3 Electrochemical Techniques

2.3.1 Open Circuit Potential Measurements (OCP)

The open circuit potential referred as the equilibrium potential, the rest potential, or the corrosion potential (E_{corr}) is the potential at which there is no current. The potential of the carbon steel electrodes immersed in the 1 M HCl was measured (against a standard saturated calomel electrode, SCE, placed in the same compartment) as a function of immersion time in the absence and presence of different concentrations from indocin-SR-expired drugs.

2.3.2 Potentiodynamic Polarization

The electrical circuit used for determining the variation of electrode potential with the electrical current. The potentiodynamic current–potential curves were recorded by changing the electrode potential automatically with a scan rate 2 mV s^{-1} from a low potential of − 800 to − 300 mV (SCE). Before each run, the working electrode was immersed in the test solution for 30 min to reach steady state. All potentials were measured against SCE. Measurements were obtained using a Voltalab 40 Potentiostat PGZ 301combined with easy corrosion program (Voltamaster 4).

2.3.3 Electrochemical Impedance Spectroscopy (EIS)

Electrochemical impedance spectroscopy [37–43] was obtained using a Voltalab 40 for all EIS measurements with a frequency range of 100 kHz–50 MHz with a 4 mV sine wave as the excitation signal at open circuit potential. If the real part is plotted on the X-axis and the imaginary part is plotted on the Y-axis of a chart, we get a Nyquist plot. The charge transfer resistance values (R_{ct}) were calculated from the difference in impedance at lower and higher frequencies [37–43].

2.4 Scanning Electron Microscopy (SEM)

Joel (840 X Japan) scanning electron microscope was utilized to document the surface morphology of various specimens of carbon steel [44]. In case of only polished surface and that immersed in 1 M HCl in the absence and presence of an inhibitor, this instrument was operated in a secondary electron imaging made with an accelerating voltage of either 10 or 20Kev. Magnifications ranging from 50 to 10000X could be obtained by this tool in the present investigation three magnifications were selected 950, 1200, and 2400X.

3 Results and Discussion

3.1 Weight Loss Measurements

3.1.1 Effect of Inhibitor Concentrations

Weight loss of C-steel in mg was calculated after fixed time 72 h in the absence and presence of different concentrations of indocin-SR-expired drugs. Figure 1 represents the relation between weight loss and immersion time. Inspection of these figures reveals that the values of weight loss increases with increasing time of immersion. The corrosion parameters such as weight loss (ΔW), rate of corrosion (k), surface coverage (θ), and inhibition efficiencies (P %) are listed in Tables 1.

The weight loss is given by the following equation:

$$\Delta W = \left(W_1 - W_2\right) \quad (1)$$

where W_1 and W_2 are the weight of specimen before and after the reaction, respectively.

In all cases, the increase of inhibitor concentrations was accompanied by a decrease in weight loss and an increase in the percentage inhibition [45]. These results lead to conclusion that the compounds under investigation are fairly efficient as inhibitors for C-steel dissolution in hydrochloric acid solution. To clear up the influence of these inhibitors on the mechanism of inhibition, the corrosion rate (k) was calculated using the following equation [46]:

$$k = \frac{\left(W_{\text{free}} - W_{\text{inh}}\right)}{At} \quad (2)$$

where k is the corrosion rate, W_{inh} and W_{free} are the weights loss of specimen in the presence and absence of inhibitor, respectively, A is the surface area in cm^2 and t is the time in hours.

The degree of surface coverage (θ) by the adsorbed molecules was calculated from equation [47]:

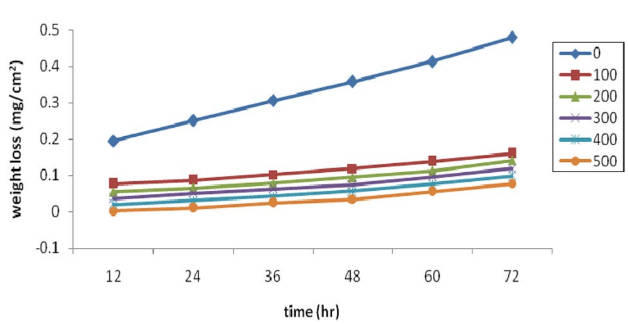


Fig. 1 Relation between weight loss and time of carbon steel in 1 M HCl in the absence and presence of different concentration of indocin-SR at 25 °C

Table 1 Effect of inhibitor (indocin-SR) concentration on carbon steel corrosion in 1 M HCl at 25 °C

Sample	Conc. (ppm)	Weight loss (mg/cm^2)	Corr. Rate (mg/cm^2 h)	θ	P (%)
Blank	0.0	0.48	0.69444	0.0	0.0
Inhibitor (indocin-SR)	100	0.1602	0.23177	0.6666	66.66
	200	0.1412	0.20428	0.7058	70.58
	300	0.1193	0.1726	0.7514	75.14
	400	0.0981	0.14193	0.7956	79.60
	500	0.0777	0.11241	0.8381	83.81

$$\theta = \frac{\left(W_{\text{free}} - W_{\text{inh}}\right)}{W_{\text{free}}} \quad (3)$$

It was found that the degree of surface coverage (θ) of the inhibitor increases by increasing the inhibitor concentration. The inhibition efficiencies (P %) of expired drugs were determined from equation [47]:

$$P\% = (\theta) \times 100 \quad (4)$$

3.1.2 Effect of Temperature

The effect of rising temperature on both corrosion and corrosion inhibition of C-steel in 1 M HCl solution at different temperatures was studied in range (25–55 °C) and was investigated by weight loss measurements. Figure 2 represent the effect of rising temperature on the inhibition efficiency obtained by weight loss method at different concentrations of the used inhibitor. The corrosion parameters such as weight loss (ΔW), rate of corrosion (k), surface coverage (θ), and inhibition efficiency (P %) for 500 ppm from compounds indocin-SR are listed in Table 2.

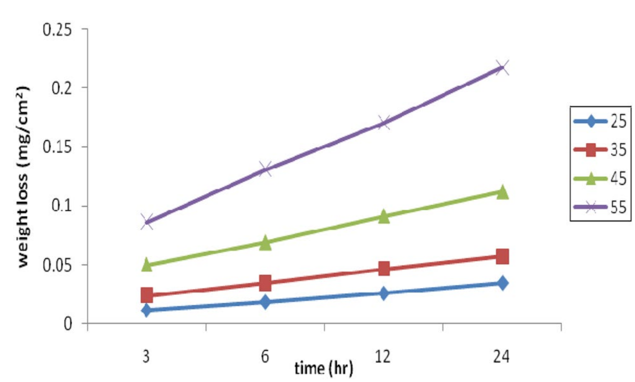


Fig. 2 Relation between weight loss and time of carbon steel in 1 M HCl in the presence of 500 ppm of inhibitor (indocin-SR) at different temperatures

Table 2 Effect of temperature on carbon steel corrosion in the presence of 500 ppm inhibitor (indocin-SR) concentration in 1 M HCl

Temp. (°C)	Weight loss (mg/cm^2)	Corr. rate (mg/cm^2 h)	θ	P (%)
25	0.0304	0.11241	0.8381	83.81
35	0.0571	0.24783	0.8125	81.3
45	0.112	0.48611	0.7921	79.2
55	0.2371	1.02908	0.7431	74.3

3.2 Kinetic Parameters

3.2.1 Activation Energy

The apparent activation energy, E_a, for the corrosion of C-steel sample in 1 M HCl solution in the absence and presence of different concentrations of expired drugs, at 25, 35, 45, and 55 °C was calculated from Arrhenius equation.

$$k = A\,e^{(-Ea/RT)} \tag{5}$$

The logarithmic form is as follows:

$$\ln k = \ln \frac{A - E_a}{RT} \tag{6}$$

where k is the corrosion rate, A is the Arrhenius constant, R is the gas constant, and T is the absolute temperature.

Arrhenius plots of ln k vs $1/T$ in the absence and presence of different concentrations of the used inhibitor are shown graphically in Fig. 3 give straight lines with slope of $(-E_a/R)$. Activation energies were calculated and given in Table 3.

The change in enthalpy and entropy of activation values (ΔH^*, ΔS^*) was calculated from the transition state theory [45].

$$k = \frac{RT}{Nh} \exp\left(\frac{\Delta S*}{R}\right) \exp\left(\frac{-\Delta H*}{RT}\right) \tag{7}$$

where h is the Plank constant, N is the Avogadro's number, and R is the ideal gas constant.

A plot of lin k versus $1/T$ gave straight lines as shown in Fig. 3a for carbon steel dissolution in 1 M HCl in the absence and presence of different concentrations of inhibitor. Straight lines are obtained with a slope of $\Delta H^*/R$ and an intercept of log $(R/\text{Nh}) + \Delta S^*/R$. The values of ΔH^* and ΔS^* are calculated and listed in Table 4.

3.2.2 Adsorption Isotherm

The type of adsorption isotherm and the effect of temperature on the corrosion rate were studied. It is generally accepted that the studied expired drug compound inhibits the corrosion process by adsorbing at the metal/solution interface [45].

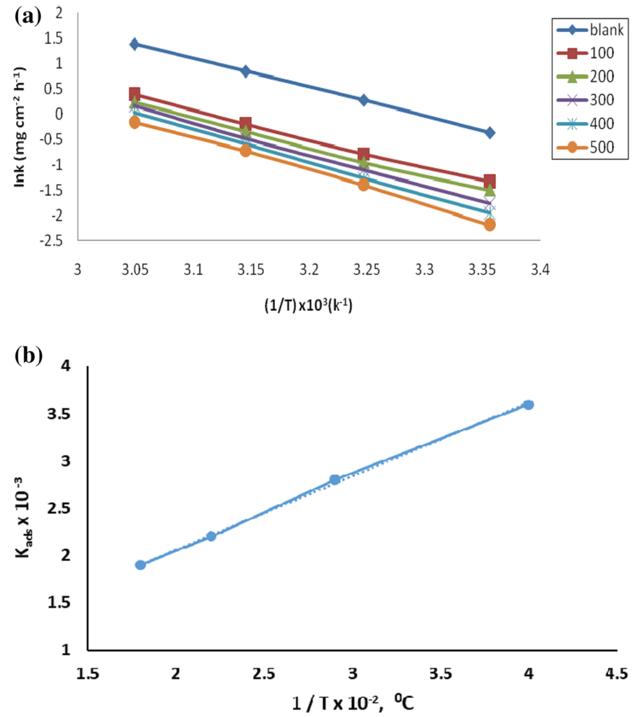


Fig. 3 **a** Arrhenius plots (ln k vs. 1/T curves) for carbon steel dissolution in the absence and presence of different concentrations of compound indocin-SR in 1 M HCl solution. **b** The relation between K ads. Vs. 1/T

Table 3 Values of activation energy (Ea*) for carbon steel in the absence and presence of different concentrations of compounds (In) in 1 M HCl at different temperatures

Conc. of inhibitor	Ea (KJ mol^{-1}) In
0.0	47.38
100	55.59
200	54.21
300	54.80
400	56.29
500	59.44

Table 4 Activation thermodynamic parameters for carbon steel in the absence and presence of 500 ppm of studied expired drugs (indocin-SR) in 1 M HCl at different temperatures

Inhibitors	ΔH^* ($KJmol^{-1}$)	ΔS^* ($J\,K^{-1}\,mol^{-1}$)
Blank	19.44	− 42.37
Compound II	24.68	− 31.42

In addition, it is believed that the formation of a solid organic molecule complex with the metal atom has received considerable attention [46].

The equation that fits our results is that due to Langmuir isotherm and is given by the general equation [33–36]:

$$\frac{C}{\theta} = \frac{1}{K_{ads}} + C \tag{8}$$

where K and C are the equilibrium constant of adsorption process and additive concentration, respectively.

The degrees of surface coverage (θ) for different concentrations of inhibitors in 1.0 M HCl solution have been calculated from weight loss measurements by using Eq. (3).

In these cases, the plots of C/θ versus C yield a straight line with intercept of ($1/K_{ads}$) and with slope, approximately equal unity was obtained, Fig. 4. The small deviation from the unity is generally attributed to the interaction of the adsorbed inhibitor molecules on heterogeneous carbon steel surface. This indicates that the adsorption of inhibitor on the carbon steel surface in 1.0 M HCl solution follows Langmuir's adsorption isotherm. The free energy of adsorption (ΔG_{ads}) at different temperatures was calculated from the following equation [45]:

$$-\Delta G_{ads} = RT \ln \left(55.5\, K_{ads}\right) \tag{9}$$

where C is the inhibitor concentration, θ is the fraction of the surface covered, K_{ads} is the equilibrium constant of the inhibitor adsorption process, the value 55.5 is the molar concentration of water in solution in mol dm^{-3}, R is the gas constant, T is the absolute temperature, and ΔG_{ads} is the standard free energy of adsorption.

The heat of adsorption (Q_{ads}) is obtained from the slopes of the straight lines when K_{ads} plot versus $1/T$, Fig. 3b, is equal to $-Q_{ads}/R$. Since the pressure is a constant, Q_{ads} is equal to enthalpy of adsorption (ΔH_{ads}) with good approximation. Entropy of inhibitor adsorption (ΔS_{ads}) can be calculated using the following equation [33–36]**:**

$$\Delta G_{ads} = \Delta H_{ads} - T\Delta S_{ads} \tag{10}$$

The calculated values of the adsorption free energy, ΔG_{ads}, adsorption entropies, ΔS_{ads}, and adsorption enthalpies, ΔH_{ads}, are given in Table 5.

3.3 Electrochemical Studies

3.3.1 Open Circuit Potential Measurements (OCP)

The potential of carbon steel electrodes immersed in 1 M HCl solution was measured as a function of immersion time in the absence and presence of different concentrations of indocin-SR drug as shown in Fig. 5. It is clear that the potential of carbon steel electrode immersed in 1MHCl solution (blank curve) tends towards more negative potential firstly, giving rise to short step. This behavior was reported [32, 48, 49] which represents the breakdown of the pre-immersion air formed oxide film presents on

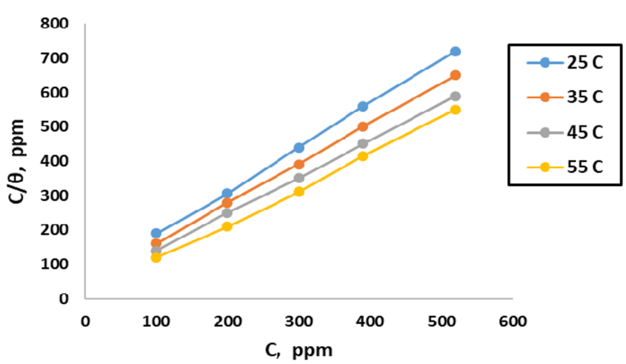


Fig. 4 Langmuir isotherm adsorption model on the carbon steel surface of compound (indocin-SR) in 1 M HCl at different temperatures

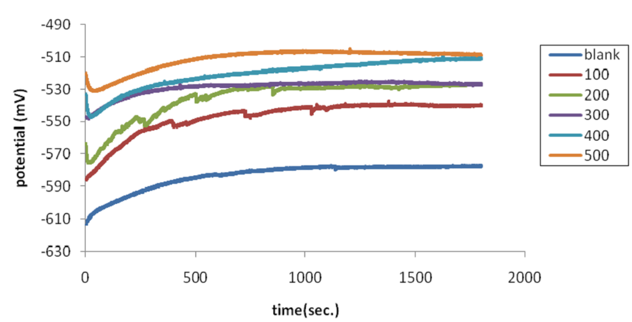


Fig. 5 Potential-–time curves for carbon steel in 1 M HCl in the absence and presence of different concentrations of inhibitor (indocin-SR) at 25 °C

Table 5 Thermodynamic parameters of adsorption on carbon steel surface in 1 M HCl containing different concentrations of compound indocin-SR at different temperatures

Temp. (°C)	K_{ads} ($\times 10^3 M^{-1}$)	ΔG_{ads} (kJ mol^{-1})	ΔH_{ads} (kJ mol^{-1})	ΔS_{ads} (J K^{-1} mol^{-1})
25	3.5	– 30.16	56.29	101.39
35	3.0	– 30.78		100.11
45	2.7	– 31.5		99.23
55	2.04	– 31.7		96.81

the surface this is followed by the growth of a new oxide film inside the solution, so that the potential was shifted again to more noble direction until steady-state potential is established. Addition of the inhibitor molecules to the aggressive medium shifts the potential to more positive direction (less negative) and as the concentration of the inhibitor increases, the corrosion potential was shifted to more noble direction [45]. In all curves, the steady-state values are always more negative than the immersion potential suggesting that before the steady-state condition is achieved, the steel oxide film has to dissolve.

3.4 Potentiodynamic Polarization Techniques

The extrapolation of anodic and/or cathodic Tafel lines of charge transfer controlled corrosion reaction gives the corrosion current density, I_{corr} at corrosion potential, E_{corr}. This method is based on the electrochemical theory of corrosion processes developed by Wagner and Traud [44]. The potential, E, is plotted as a function of logarithm of current density (I) and shows the polarization curves for both anodic and cathodic reactions [49–53].

Figure 6 represents the potentiodynamic polarization plots for carbon steel electrode in 1 M HCl in the absence and presence of different concentrations of the inhibitor, at scanning rate 2 mV/sec. The percentage inhibition efficiency P % is given by:

$$P\,\% = \left\{ \frac{\left(I_{corr} - I_{corr(inh)}\right)}{I_{corr}} \right\} \times 100 \quad (11)$$

where I_{corr} and $I_{corr(inh)}$ are the corrosion current densities in the absence and presence of inhibitors, respectively, determined by extrapolation of cathodic Tafel lines to the corrosion potential.

The degree of surface coverage (θ) was calculated by the following equation [54]:

$$\theta = \left\{ \frac{\left(I_{corr} - I_{corr\ (inh)}\right)}{I_{corr}} \right\} \quad (12)$$

Table 6 shows the effect of inhibitor concentration on some electrochemical parameters, corrosion current density (I_{cror}), corrosion potential (E_{corr}), anodic and cathodic Tafel slopes (β_a and β_c), the degree of surface coverage (θ), and the percentage of inhibition efficiency (P %) during the corrosion of C-steel electrode in 1 M HCl solutions.

3.5 Electrochemical Impedance Spectroscopy (EIS)

The corrosion behavior of C-steel in 1.0 M HCl solution in the absence and presence of the used expired drug (indocin-SR) was investigated by EIS method at 25 °C. Figure 7 show the Nyquist plots for carbon steel in 1 M HCl solution in the absence and presence of different concentrations of inhibitors at 25 °C. The Nyquist plots were

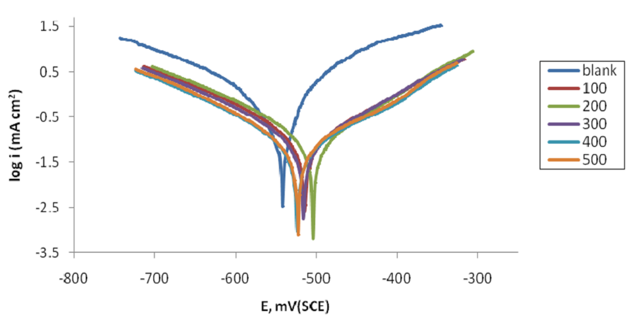


Fig. 6 Potentiodynamic polarization curves for the carbon steel in 1 M HCl in the absence and presence of different concentrations of compound (In-SR) at scanning rate 2 mV s^{-1}

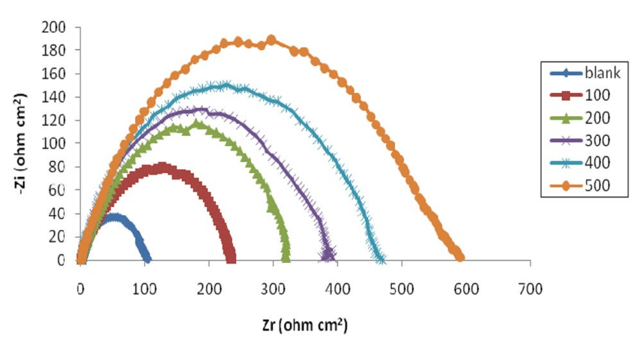


Fig. 7 Nyquist plots for the carbon steel in 1 M HCl in the absence and presence of different concentrations of compound indocin-SR

Table 6 Potentiodynamic polarization parameters for corrosion of carbon steel in 1 M HCl in the absence and presence of different concentrations of compound indocin-SR at 25 °C at scanning rate 2mVs^{-1}

Conc. of inhibitor	E_{corr} (mV)	Icorr (mA cm^{-2})	Ba (mV dec^{-1})	Bc (mV dec^{-1})	θ	P (%)
Blank	− 542.2	0.4622	154.9	− 319.6	–	–
100	− 514.4	0.1918	132.4	− 149.8	0.5850	58.5
200	− 504.3	0.1783	117.4	− 146.4	0.6142	61.42
300	− 516.0	0.1412	121.9	− 138.7	0.6945	69.45
400	− 523.6	0.1065	123.0	− 132.8	0.7695	76.95
500	− 522.8	0.0942	114.3	− 122.1	0.7961	79.61

Table 7 EIS parameters for corrosion of carbon steel in 1 M HCl in the absence and presence of different concentrations of compound indocin-SR at 25 °C

Conc. of inhibitor	Rct (Ohm cm^2)	Rs (Ohm cm^2)	Cdl (μF cm^{-2})	θ	P (%)
Blank	104.269	2.18	40.95	–	–
100	237.012	1.64	8.56	0.5600	56.00
200	322.789	1.58	4.19	0.6769	67.69
300	392.478	2.23	3.13	0.7343	73.43
400	470.124	1.86	2.25	0.7782	77.82
500	591.568	1.74	1.42	0.8237	82.37

regarded as one part of a semicircle. The impedance diagram shows the same trend (one capacitive loop); however, the diameter of this capacitive loop increases with increasing concentration. The presence of the inhibitor increases the impedance but does not change other aspects of the behavior. These results support the results of polarization measurements that the inhibitor does not alter the electrochemical reactions responsible for corrosion. It inhibits corrosion primarily through its adsorption on the metal surface [55, 56]. The charge transfer resistance values (R_{ct}) are calculated from the difference in impedance at lower and higher frequencies, as suggested by Haruyama and Tsuru [30]. To obtain the double-layer capacitance (C_{dl}), the frequency at which the imaginary component of the impedance at maximum $f(-Z''_{img})$ is found and C_{dl} values are calculated from the following equation [30, 57]**:**

$$f\left(-Z''_{\text{img}}\right) = 1/\left(2\pi C_{\text{dl}} R_{\text{ct}}\right) \quad (13)$$

The impedance quantitative results can be seen in Table 7. It is clear that the corrosion of steel is obviously inhibited in the presence of the inhibitor. It is apparent that the impedance response for carbon steel in 1 M HCl changes significantly with increasing inhibitor concentration [58–60].

In the case of the electrochemical impedance spectroscopy, the inhibition efficiency is calculated using charge transfer resistance as follows [37–43]**:**

$$P\% = \left\{\left(R_{\text{ct(inh)}}\right) - R_{\text{ct}})/R_{\text{ct(inh)}}\right\} \times 100 \quad (14)$$

where R_{ct} and $R_{ct(inh)}$ are the charge transfer resistance values in the absence and presence of inhibitors for C-steel in 1 M HCl, respectively. As the inhibitor concentration increased, the R_{ct} values increased, but the C_{dl} values tended to decrease. The decrease in C_{dl} value is due to the adsorption of inhibitor on the metal surface [61]. The inhibition efficiency increases with increasing inhibitor concentration. This fact suggests that the inhibitor molecules may first be adsorbed on the steel surface and cover some sites of the electrode surface. These layers protect steel surface from corrosion [37–43]**.**

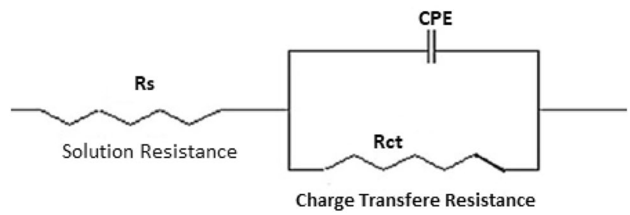


Fig. 8 The equivalent circuit model for the electrochemical impedance measurements

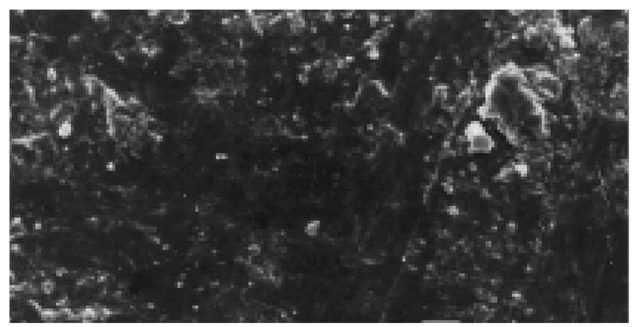


Fig. 9 Scanning electron micrographs of carbon steel sample after polishing

The equivalent circuit Fig. 8 consists of the double-layer capacitance (C_{dl}) in parallel to the charge transfer resistance (R_t) which is in series to the parallel inductive (Rs) [54, 58–60].

3.6 Scanning Electron Microscopy (SEM)

The scanning electron microscope (SEM) is a type of electron microscope that images the sample surface by scanning it with a high energy beam of electrons in a raster scan pattern. The electrons interact with the atoms (at or near the surface of the sample) that make up the sample producing signals that contain information about the sample's surface topography [58]. Figure 9 shows a characteristic inclusion observed on the polished carbon steel surface, which was probably an oxide inclusion, so that a comparison can be drawn with the morphology after exposure to the corrosive media. Figure 10 shows SEM image of the surface of carbon

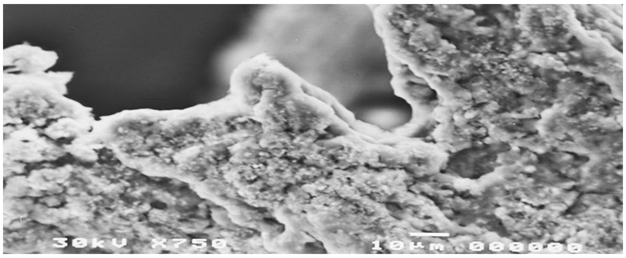


Fig. 10 Scanning electron micrographs of carbon steel sample after immersion in 1 M HCl solution without inhibitor

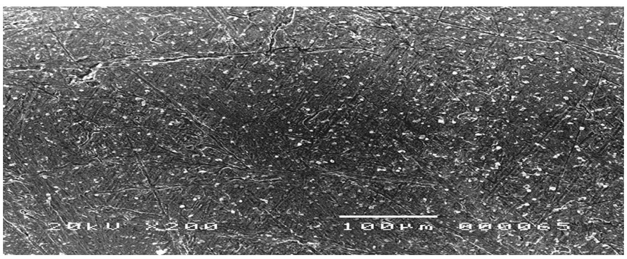


Fig. 11 Scanning electron micrographs of carbon steel sample after immersion in 1 M HCl solution containing 500 ppm of inhibitor (indocin-SR)

steel specimen after immersion in 1 M HCl for 24 h, while Fig. 11 shows SEM image of another carbon steel specimen after immersion in 1 M HCl for the same time interval in the presence of 500 ppm of the inhibitors. SEM observations of the steel surface showed that a film of inhibitor molecules is formed on the electrode surface, this film retarded both the reduction of hydrogen ions (cathodic reaction) and the anodic dissolution of carbon steel (anodic reaction).

4 Conclusion

Based on the obtained results, the following conclusions are accomplished: -

1- The use of the expired drugs as corrosion inhibitors is of high advantages, like (a) it low price, (b) high availability, (c) save, and eco-friendly, (d) nontoxic inhibitors
2- The studied expired drugs, indocin–SR capsules, are excellent inhibitors and act as the mixed-type inhibitors for carbon steel corrosion in hydrochloric acid solution.
3- Inhibition efficiencies increased by increasing inhibitor concentration and decrease by increasing temperatures up to 328 K.
4- The uniform increasing inhibition efficiency as the function of concentration and the adsorption of all additives obeys the Langmuir adsorption isotherm.
5- All entropy parameters for adsorption of inhibitor molecules on carbon steel are negative and increased by increasing the temperature which indicates that the inhibitor being more oriented and less ordered on the surface of the metal.
6- The activation parameters of the adsorption (E^*, ΔH^* and ΔS^*) were calculated and showed that the used inhibitors decreased the rate of corrosion.
7- SEM observations of the steel surface showed that a film of inhibitor molecules is formed the electrode surface; this film retarded both the anodic and cathodic reactions.

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