

The Corrosion Inhibition of The Carbon Steel and Oil Pipelines in 1 M H₂SO₄ by Expired Atarax and Tussinor, Drugs

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Abstract: Metal corrosion is an industrial and economic problem, especially in the oil industry, where it is transported and stored in carbon steel tanks. In turn, we are looking for inexpensive, economical and effective solutions to increase industrial development and economic growth. We have studied two expired drugs, Atarax. (At.) (Hydroxyzine hydrochloride), and Tussinor (Tu.) drugs. It has been found that the efficiency of (Tu) is higher than (At), especially at low concentrations of 100 ppm, whether by weight loss method or by Potentiometric method, and that it also proved the highest efficiency at high temperatures, which was also confirmed by the Arrhenius curves and the transition state. Potentiometric method also showed a higher stability of (Tu.) than (At.) by measuring the voltage within two hours, and the voltage also increased with the increase in the concentration of the expired drugs, and the current density decreased with the increase in concentration and increased with the increase in potential. As indicated by the Arrhenius curves, the transition state, the calculation of the activation energy, the change in enthalpy, and the change in entropy, the process of corrosion and inhibition are monitored by the change in voltage using a voltage and current meter and by using Ag/AgCl as a reference electrode.

Keywords: Corrosion inhibitors, Carbon steel, Oil pipelines, Expired drugs, Potentiometric method.

Introduction:

Corrosion is defined as the interaction of a metal with its surrounding environment of gases and liquids. Expired drugs are good fluids for corrosion and are used as corrosion inhibitors (CI) and to limit the interaction of the metal with its environment. Therefore, drugs or chemical compounds are considered environmentally friendly alternatives to (CI) for use in various industrial and biological applications. Not only this, the cost of re-equipment eroding in the industry negatively affects the economy. The use of expired drugs as (CI) are a low-cost, cost-effective, and environmentally friendly alternative to the high-cost corrosion process. For example, used an expired Melatonin drug used as (CI) of carbon steel, (CS) in 0.5M H₂SO₄ solution [1], natural ligands including amino acids, natural polymers like Proteins, and Carbohydrates), Polyphenols, Flavonoids, Fatty acids, Oleochemicals, Purines and Pyrimidines, Drugs and Phytochemicals were used as (CI) [2], an expired (Amoxicillin, Ciprofloxacin, Doxycycline and Streptomycin,) which classes as antibacterial drugs were used as (CI) for steel in aqueous solution (pH 4) simulating an acid rain [3], an expired Penicillin G, Ampicillin and Amoxicillin drugs were used as (CI) of (CS) (ASTM 1015) in 1M HCl solution [4], an expired Irbesartan drug used as (CI) on mild steel (MS) corrosion in 1M

HCl and 0.5M H₂SO₄ solutions [5], an expired Antihypertensive drugs were used as (CI) of Al and three Al-Si alloys in different concentrations of HCl solutions [6], an expired Ondansetron HCl (ODSH) drug was used as the inhibiting action against the corrosion of (MS) in acid medium [7], an expired Streptomycin was used as (CI) of (MS) in 1M HCl solution [8], an expired Clotrimazole (CTM) and Fluconazole (FLC), two antifungal drugs, were used as (CI) for Al in 0.1 M HCl solution [9], an expired Cefalexin was used as (CI) of (MS) in 1N HCl solution [10], an expired Bupropion was used as (CI) of (CS) in 1M HCl and 0.5M H₂SO₄ solutions [11], an expired Lansoprazole and Rabeprazole on the corrosion protection of (CS) in 3.0 M H₃PO₄ solution [12], four compounds of Antibacterial drug were used as (CI) of Al in 2M HCl solution in the absence and presence of drugs [13], Isoniazid with 5-substitute Indole derivatives were synthesized and investigated for (CI) of (MS) in 1M HCl medium [14], an expired Salbutamol drug as an emerging anticorrosion additive for (MS) corrosion in oilfield acidizing fluid [15], Cefotaxime Sodium drug (Cefo) is avoiding Al corrosion in alkaline media as Al (CI) in 0.1 M NaOH [16], Linagliptin (LGP) was used as (CI) of a (MS) in 1M HCl solution [17], an expired Paracetamol was used as (CI) of copper in a synthetic acid rain solution [18], Acetylsalicylic acid (ASA) molecules was used as

(CI) of (CS) XC48 in 1M HCl solution [19], Isoxsuprine HCl drug was used as (CI) of (MS) corrosion in a 1M HCl solution [20], organic compounds were used as (CI) for stainless steel (SS) in various aggressive environments such as H₂SO₄, HCl, H₃PO₄, sulphamic acid, CO₂, H₂S, and NaCl at different temperatures, in acid medium [21], Tragacanth Gum (TG) and Ceftriaxone (Cef) were used as (CI) of (MS) in HCl solution [22], Three Oxazolone derivatives as potential (CI) were used as (CI) of API5LX60 graded (CS) in 1N H₂SO₄ solution at concentrations 50–200 ppm of drugs [23], Urispas drug was used as (CI) of commercial (MS) in 1M H₂SO₄ solution [24], Guava leaf extract was used as a green (CI) in biodiesel [25], Benzoyl Thiourea, drug was used as (CI) of commercial (MS) in 1M H₂SO₄ solution [26], Methyl Carbazodithoate was used as (CI) of commercial (MS) in 2M H₂SO₄ solution [27]. In this work an expired Atarax (At.), Hydroxyzine hydrochloride and Tussinor (Tu) drugs were used as (CI) for (CS) in 1M H₂SO₄ solution has been studied by weight loss, and potentiometric method.

Experimental

2.1. Materials and Chemicals

The carbon steel sheets with the following chemical composition (wt%): C (0.200g), Si (0.003g), Mn (0.35g), P (0.02g) and Fe (Rest). The piece area was $1.23 \times 10^{-3} \text{ m}^2$. The sample was embedded in a glass tube of just larger diameter than the sample. Epoxy resin (supplied from Ciba Co.) was used to stick the sample to the glass tube. Surface of carbon steel electrode was mechanically rub off using sand papers, in different grades, for example 1200 grade, before used. The tests were used 1M H₂SO₄ (supplied from Sigma-Aldrich) with the addition of various concentrations of nature products compounds. All the test solutions were prepared from analytical chemistry grade chemical reagents prepared using distilled water, and used without further pure cation. For each hold, a freshly prepared solution was used. Temperature of solutions was thermostatically controlled at desired value, and all chemical material high purity about 99%. [28]

2.2 Apparatus

All Potentiometric measurements were made at $25 \pm 1^\circ\text{C}$ with an Orion (Model 720) pH/mV meter (Fisher scientific). Double junction Ag/AgCl reference electrode was used with Digital multimeter (TMT480012). All chemicals were of analytical reagent grade unless otherwise stated and distilled water was used throughout. Testing was performed using dielectrode electrochemical cell with a volume of 250 ml. The working electrode was made of the steel with an exposed to solution area of 0.00123 m^2 , the reference

electrode was Ag/AgCl electrode, The inhibitors were added into the test solution. The current recorder by mA, the potential mV, and the time by min. The inhibitor measurements, the potential vs time dependences of the steel in 1M H₂SO₄ solution without and with the addition of an expired drugs as inhibitors were recorded.

2.3 Tools and working method:

Voltage and current multimeter device, a glass beaker with a capacity of 250 ml, 1M H₂SO₄ titrated with Na₂CO₃ of accurate concentration, Ag/AgCl as a reference electrode prepared by dipping two silver electrodes in a 1M solution of HCl and passing a direct current between them from a 1.5 Volt battery The working electrode is (CS) electrode.

2.4 Cell preparation:

The cell consists of two electrodes, one of which is Ag/AgCl electrode as a reference electrode, while the working electrode is the (CS) electrode, and an area immersed in acid $1.23 \times 10^{-3} \text{ m}^2$.

Voltage (mV) and current (mA) are measured every ten minutes for 2 hours, during which corrosion of (CS) takes place in the presence of 1M H₂SO₄ acid. Voltage, current and time are measured in the presence of different concentrations of (At.& Tu.) an expired drugs were used as (CI) at concentrations of 100, 200, 300, 400, ppm and for a 2 hour for each concentration.

2.5. Weight loss measurements

Experiments were performed with different concentrations of (CI). The immersion time for the weight loss is 2 h at 25°C . The results of the weight loss experiments are the mean of three runs, each with a fresh specimen and 100 ml of fresh acid solution. The inhibition efficiency $IE_w\%$ and $IE_e\%$ were calculated.

Weight loss calculations are comprehensive corrosion tests for laboratory and field. Also, they help us to make a quantitative estimate of amount of corrosion. The corrosion behaviour of the metal in an aqueous environment is describe by the extent to which it dissolves in the water solution. Calculated the weight of a specimen before and after precipitate and applying the following equation:

$$W = \frac{m_1 - m_2}{At} \text{ ----- (1)}$$

where m_1 and m_2 – the mass of the sample before and after testing, respectively, g; A – area of the sample, m^2 ; t– exposure time, hours.

$$IE_w\% = \frac{W_0 - W}{W_0} \times 100 \text{ ----- (2)}$$

where W_0 and W – corrosion rate of steel in test solution without and with (CI), respectively.

All Potentiometric measurements were made at $25 \pm 1^\circ\text{C}$ with an Orion (Model 720) pH/mV meter (Fisher scientific). Double junction Ag/AgCl

reference electrode was used with Digital multimeter (TMT480012). All chemicals were of analytical reagent grade unless otherwise stated and distilled water was used throughout. Testing was performed using dielectrode electrochemical cell with a volume of 250 ml. The working electrode was made of the steel with an exposed to solution area of 0.001 m², the reference electrode was silver chloride electrode, (CI) were added into the test solution. The effect of inhibition IE_e was determined by the formula:

$$IE_e = \frac{I_0 - I_{inh}}{I_0} \times 100 \quad (3)$$

where I_0 and I_{inh} - corrosion current density of steel in test solution without and with inhibitor, respectively. The current recorder by mA, the potential mV, and the time by min.

Expired drug measurements, the potential vs time dependences of the (CS) in 1M H₂SO₄ solution without and with the addition of inhibitors were recorded.

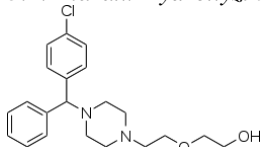
2.6. Preparation of expired drug solution

The structure, nomenclature, molar mass, & molecular formula of expired drugs one liter stock solution (1000 ppm) of an investigated antibiotic, as tablets, was prepared by dissolving an accurately weighed quantity (1g) in (1L) of doubly distilled H₂O, and then the desired concentrations (100 ppm - 400 ppm) are obtained by diluting the stock solution with the desired volume of distilled H₂O. The corrosive solution, 1M H₂SO₄ was prepared by diluting of high-grade H₂SO₄ (98 % w, d = 1.84 g/ml, M. wt.= 98 g/mol.) with distilled water and dilute to up to 1M then titrated with 1M Na₂CO₃ standard solution to obtain exact 1M H₂SO₄ at suitable indicator.

(CS) sample has area 1.23×10^{-3} m² was placed in 1M H₂SO₄ in a beaker devoid of, and with distinct quantities of the drugs for 2 hours at 298-323 K. Then they washed, desiccated, weighed after 10 min and measure the current and potential.

Results and discussion

3.1. Atarax. Hydroxyzine hydrochloride (At)



(±)-2-(2-[[4-[(4-chlorophenyl)-phenylmethyl]piperazin-1-yl]ethoxy)ethanol

3.2. Tussinor drug (Tu) Scientific Name: Guava leaves ext.+ Tilia ext.+ Fennel oil+ Pure Honey, Guava leaves ext. contains tannins, phenols, flavonoids, terpenoids, and glycosides [30].

Chemical drugs (drugs) were used as (CI) of metal or steel corrosion is one of the most effective, green corrosion, environmentally friendly, and

cheap things. This is due to its composition diversity, complex structures and large surface area, The effective different functional groups in the drug molecules provide good coverage and protection for the surface. To explain how the drug molecules act as (CI) where the drug molecules provide excellent surface coverage and protection as its molecules contain many electrons rich centers such as polar functional groups and aromatic rings through which they adsorb and act effectively as excellent (CI). The use of chemical drugs (drugs) as inhibitors of metal corrosion is one of the most effective, environmentally friendly and cheap things. This is due to its complex structures and large surface area. The effective functional groups in the drug molecules provide good coverage and protection for the surface. [31].

The corrosion rate and inhibition efficiency for steel in 1M H₂SO₄ solution at 25°C in the absence and presence of (At. &Tu.) drugs are given in **Fig. 1**. As the (At. &Tu.) expired drugs increase, the corrosion rate decreases. In other words, the inhibition efficiency of (At. &Tu.) expired drug increases with the increase of its concentration to attain 95%.

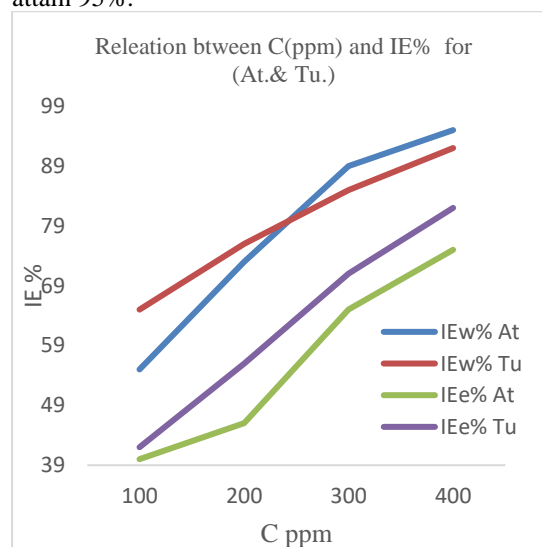


Fig.1. Effect of (At. &Tu.) expired drugs concentrations (ppm) in corrosion inhibition efficiency of (CS) in 1M H₂SO₄ solution.

In Fig.1. Shows that, as the concentration of inhibitors increases the inhibition efficiency also increases, due to the numbers of molecules were adsorbed on the surface of (CS), also increases.

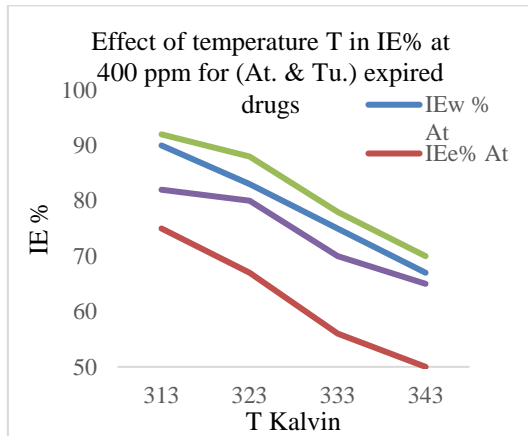


Fig.2. Effect of T Kalvin on IE% at 400 ppm for (At. &Tu.) expired drugs.

In Fig.2. Shows that as increase in temperature decrease the efficiency due to increase the kinetic energy of molecules of drugs adsorbed, then decrease the numbers of molecules were adsorbed on the surface of (CS). It clearly indicates that temperature mainly influences the desorption of adsorbed inhibitor on the (CS) surface. As the concentration of inhibitors increases and inhibition efficiency also increases, but when the temperature increases the inhibitor efficiency decreases this is because at a high temperature, the hydrogen evolution increases on the metal surface and desorption of the thin film takes place and so inhibitor efficiency decreases. When the temperature increases the corrosion rate is increased linearly, it is clearly indicates that temperature mainly influences the desorption of adsorbed inhibitor on the metal surface.

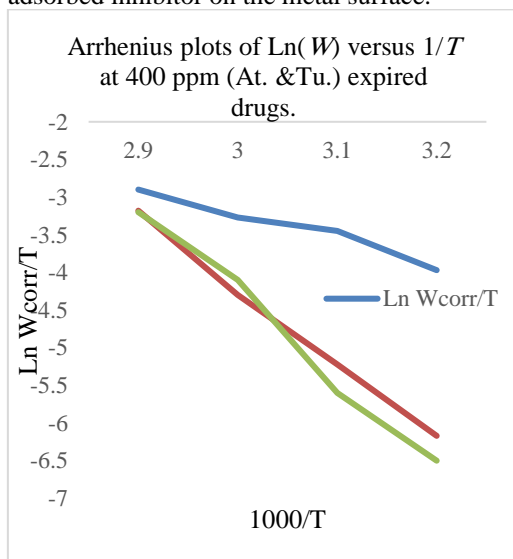


Figure 3. Arrhenius plots of Ln(W) vs 1/T at 400 ppm for (At. &Tu.) expired drugs.

In Fig.3. Shows the relation bet and Ln(W) as Arrhenius equation (4) to calculate the activation energy (Ea) for the corrosion of (CS) in the absence

and presence of 400 ppm concentrations of (Ch. & Rh.), Arrhenius-type equation:

$$W_{corr} = Ae^{\frac{-E_a}{RT}} \text{ --- (4)}$$

where (Ea) is the activation corrosion energy; R is the universal gas constant; A is the Arrhenius pre-exponential factor, T is the absolute temperature and Wcorr is corrosion rate, and from Arrhenius plots for the corrosion rate of (CS) in 1M H₂SO₄, the values of Ea were evaluated from the slope of ln W versus 1/T plots.

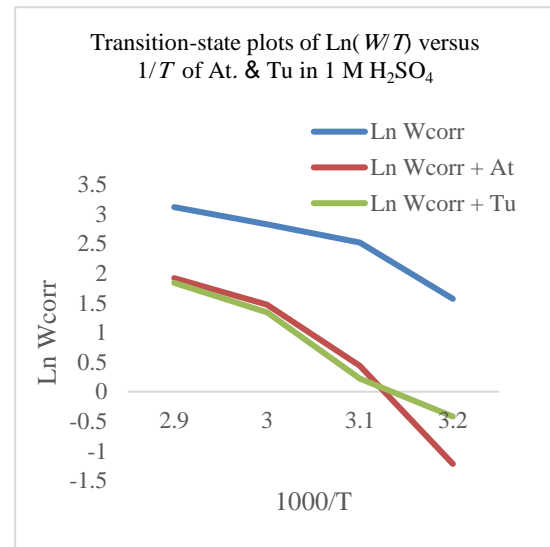


Fig. 4. Transition-state plots of Ln(W/T) vs 1/T in 1M H₂SO₄ in absence and presence of 400 ppm (At. &Tu.) expired drugs.

In Fig. 4. Shows the relation between 1/T and Ln(W/T) for (At. &Tu.) expired drugs, in 1M H₂SO₄ to calculate, the enthalpy of activation (ΔH*) and the entropy of activation (ΔS*) for the corrosion of (CS) in H₂SO₄ may be estimated using the transition-state equation:

$$W_{corr} = \frac{K_B T}{h} \exp\left(\frac{\Delta S^*}{R}\right) \exp\left(-\frac{\Delta H^*}{RT}\right) \text{ --- (5)}$$

Where, k_B is the Boltzmann's constant and h is the Planck's constant. A plot of ln(W/T) versus 1/T, straight lines was obtained with a slope of -ΔH* and from the intercepts of log(W/T)-axis, ΔS* values were calculated.

The data was collected in Table.1 indicate that the addition of (At. & Tu.) leads to an increase in the activation Ea and ΔH* to values greater than that of the free solution. Moreover, the average difference value of the Ea - ΔH* is 2.6 kJ/mol which is approximately equal to the value of RT (2.63 kJ/mol) at the average temperature (238 K) of the domain studied. This result agrees that the corrosion process is a unimolecular reaction as described by the known equation of perfect gas [29].

$$E_a - \Delta H^* = RT \text{ --- (6)}$$

It is pointed out in the literature that positive sign of the enthalpies reflects the endothermic nature of the (CS) dissolution process, the presence of inhibitors tested reveals that the corrosion process becomes more and more endothermic when compared to blank.

Table 1. Shows that, E_a , ΔH°_{ads} , and ΔS°_{ads} for the corrosion of (CS) in 1M H_2SO_4 in the absence and presence of (At. & Tu.) expired drugs.

	E_a (kJ/mol)	ΔH°_{ads} (kJ/mol)	ΔS°_{ads} (J/mol.K)
Blank	35	32.4	- 33.42
At	85.4	82.6	- 83.6
Tu	84.45	81.2	-82.6

The activation energies are $E^*_a = 35 \text{ kJ mol}^{-1}$, $E^*_a = 85.4 \text{ kJ mol}^{-1}$ for At., and $84.45 \text{ kJ mol}^{-1}$ for Yu were the activation energies in the absence and presence of drugs, respectively. The low value of the activation energy (less than 80 kJ mol^{-1}) indicate physical adsorption of the drugs molecules on metal surface.

The decrease of % IE with temperature is explained by the adsorption of an organic adsorbate on the surface of a metal is regarded as a substitutional adsorption process between the organic compound in the aqueous phase, org_{aq} , and the water molecules adsorbed on the electrode surface H_2O_{suf} .

Large and negative values of entropies show that the activated complex in the rate determining step represents an association rather than a dissociation step, meaning that a decrease in disordering takes place on going from reactants to the activated complex.

3.3. Potentiometric results

The activity of specific cations and anions can measure potentiometry with ion-selective microelectrodes at the solid/liquid interface from the solution side. Anodic and cathodic components of corrosion processes change the concentration of H^+ , OH^- in local solution, ions of the supporting electrolyte, and metal cations [32].

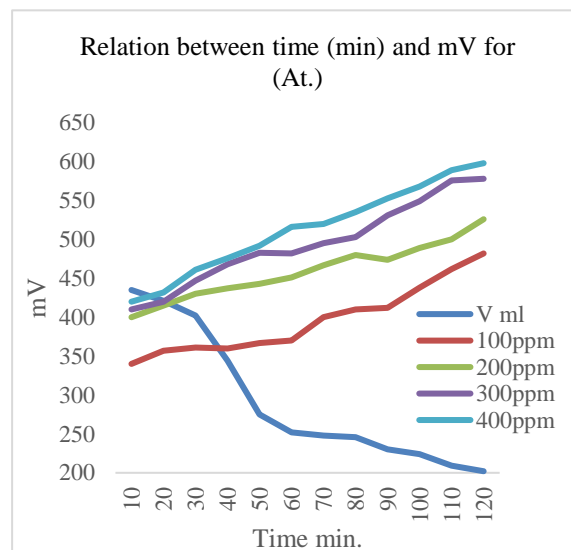


Fig. (5). Relation between time (min.) and mV at different concentration of expired (At.) drug.

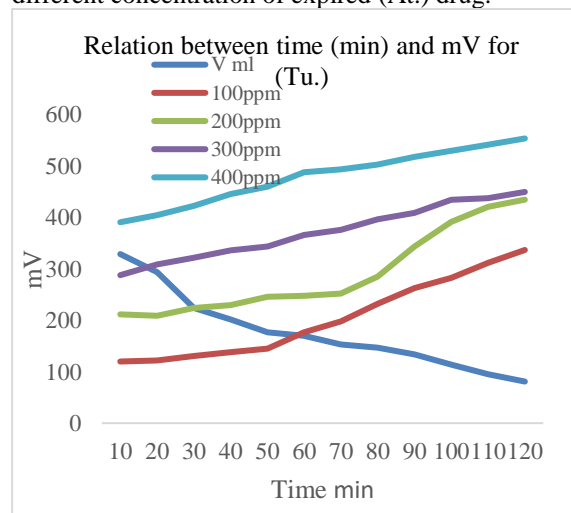


Fig.6. Relation between time (min) and mV at different concentration of expired (Tu.) drug.

In Fig. 5,6 shows that the voltage during the corrosion process without adding (At. & Tu.) expired drugs, which is used as (CI) for (CS) in the presence of H_2SO_4 , we find that the voltage decreases with the increase in corrosion processes, and with the increase of time the voltage decreases to increase the formation of a layer of oxide that increases the conductivity of the solution. Then the voltage decreases between the working electrode and the reference electrode, while the voltage increases when (At. & Tu.) drugs were added as a result of adsorption of the drug on the surface of (CS), which makes a protective layer between (CS) and the solution, and it increases gradually with increasing time until two hours, and the voltage increases as the concentration of expired (At. & Tu.) drugs increases from a concentration of 100 ppm to 400 ppm.

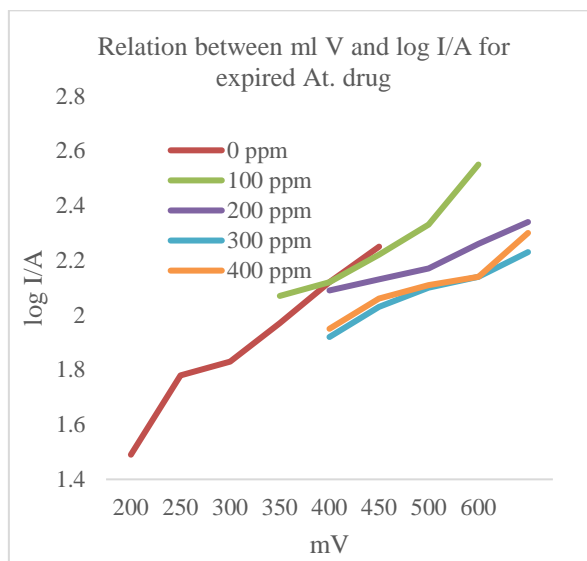


Fig.7. Relation between mV and log I/A at different concentration of expired (At.) drug.

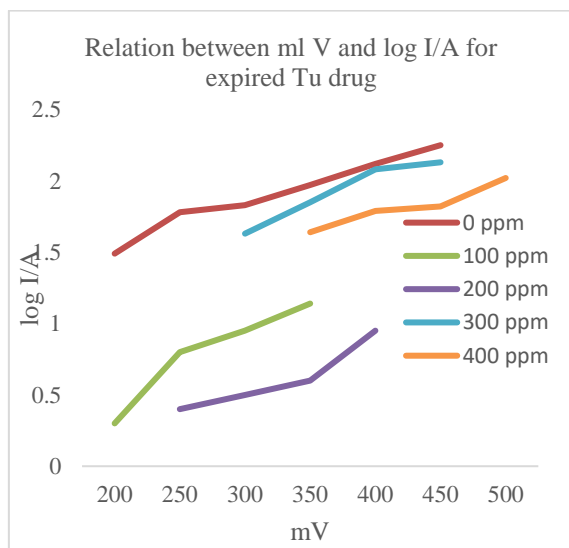


Fig. 8. Relation between mV and log I/A at different concentration of expired (Tu.) drug.

In Fig. 7. Shows the relationship between the voltage and the logarithm of the current density, which is the current intensity in amperes per m^2 . We note that the current density is higher when the solution is without an inhibitor (0 ppm), then it decreases as the concentration of the inhibitor increases at the same voltage, for example at a voltage of 400 mV, we find that the density decreases. The current increases with the concentration of the inhibitor, although it generally increases with the increase in the voltage, because the current generally increases with the increase in the voltage, because the current generally increases with the increase in the voltage.

It was noted here that the voltage increase is delayed at a concentration of 100 ppm and its

decrease with time may be due to the fact that the layer formed at this concentration did not adhere sufficiently or because the drug contained more than one compound (three compounds) that were not all of the same degree of efficiency in the process of adsorption or inhibition corrosion process, but it began to increase its efficiency by increasing the concentration up to 400 ppm.

In Fig. 8. Shows that the current density at the same voltage is 450 mV for a concentration of 100 ppm less than the current density of the solutions without (CI) and the high voltage and they converge together at a concentration of 200 ppm while the voltage increases significantly at higher concentrations and we could not measure the current density of the solution Inhibitor-free, because the cell voltage did not reach these high values except in the presence of high concentrations of the inhibitor.

Conclusion

Tussinor drug (Tu) is more efficient than Atarax. Hydroxyzine hydrochloride (At), by weight loss method or by Potentiometric, and its more stable at high temperatures, this was confirmed by the Arrhenius curves, state and transition, activation energy calculation, change in enthalpy, change in entropy, and the voltage method showed the extent of voltage change with time for each, concentration of expired drug for two hours, that the voltage increases with increasing concentration, which indicates a high ability of the inhibitor to stick to the metal.

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تشبيط تآكل الفولاذ الكربوني وخطوط أنابيب النفط في 1 مولار من حمض الكبريتيك باستخدام أدوية أتراكس وتسنور منتهية الصلاحية.

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الملخص

يمثل تآكل المعادن مشكلة صناعية واقتصادية وخاصة في مجال صناعة النفط حيث ينقل ويخزن في خزانات من الصلب الكربوني وبدورنا نبحث عن حلول غير مكلفة اقتصادية وناجعة لزيادة التطور الصناعي والنمو الاقتصادي. وقد قمنا بدراسة دوائين منتهيا الصلاحية هما أتراكس وتوسنيور وقد وجد أن كفاءة توسنيور اعلي من أتراكس خاصة عند التركيزات المنخفضة ١٠٠ جزء في المليون سواء بطريقة فقد الوزن او بالطريقة الجهدية وأنه أثبت ايضا وأعلي كفاءة عند درجات الحرارة المرتفعة وهو ما أكدته أيضا منحنيات أرهينيوس والحالة الانتقالية. وقد أظهرت ايضا الطريقة الجهدية ثباتا اعلي لدواء توسنيور عن أتراكس من خلال قياس الجهد خلال ساعتين ويزداد الجهد ايضا بزيادة تركيز الدواء منتهي الصلاحية وتنخفض الكثافة التيارية بزيادة التركيز وتزداد بزيادة الجهد. كما دلت على ذلك منحنيات ارهينيوس والحالة الانتقالية وحساب طاقة التنشيط والتغير في المحتوي الحراري والتغير في الإنتروبي وقد تم متابعة عملية التآكل والتشبيط بواسطة التغير في الجهد باستخدام مقياس الجهد والتيار وباستخدام جهد الفضة وكلوريد الفضة كقطب مرجعي.

الكلمات المفتاحية : أدوية منتهية الصلاحية، الصلب الكربوني، الطريقة الجهدية، أنابيب النفط، مثبتات التآكل.