The Corrosion Inhibition of The Carbon Steel and Oil Pipelines in 1M HCl by Expired Lactulax and Triazepine Carboxylate Drugs

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Abstract: Corrosion in carbon steel and oil pipelines has adverse economic and environmental effects. Therefore, there is interest in utilizing inexpensive and environmentally friendly materials to prevent and inhibit corrosion. In this study, two expired drugs, Lactulose (La.) and Triazepine Carboxylate (TC), were investigated as potential inhibitors of steel corrosion in 1M HCl. The weight loss method and the potentiometric method were employed to calculate the inhibition efficiency (IEw%) of these compounds. The study examined the effect of inhibitor concentration and temperature on IEw%, and also involved calculating the activation energy and enthalpy change using the Arrhenius equation and transition state theory, according to the weight loss method, La. exhibits higher inhibition efficiency than TC. However, both inhibitors show nearly equal efficiency according to the potentiometric method. La. demonstrates greater stability and efficiency at elevated temperatures, as evidenced by Arrhenius plots and transition state analysis, which also involved calculating activation energy, enthalpy change, and entropy change. The corrosion process and inhibition were monitored by changes in voltage using a voltage and current meter, with Ag/AgCl used as a reference electrode. It was observed that La. exhibits shorter adsorption stability on the surface of carbon steel compared to TC, which contributes to its higher efficiency in inhibiting corrosion.

Keywords: Corrosion Inhibitors - Carbon Steel - Oil Pipelines - Expired Drugs - Potentiometric Method- Lactulose -Triazepine Carboxylate.

Introduction:

Metal surfaces react with gases and fluids, leading to their destruction through environmental interactions. As a result, chemical medicines or drugs are being explored as environmentally friendly corrosion inhibitors (CIs) in various industrial and biological applications. The economic repercussions of repairing equipment damaged by corrosion are significant. Using expired drugs as CIs offers a cost-effective and eco-friendly alternative to expensive degradation processes, crucially protecting metals and alloys with implications for technology, economics, and the environment.

Expired antiplatelet drugs have been used as CIs for 316L stainless steel (SS) in simulated body fluid [1], while expired Rhodanine Azosulpha drugs were employed as CIs for 304 SS in 1M HCl solution [2]. Additionally, expired Vitamin B1 (VB1), containing a thiazole ring, was initially used as an eco-friendly inhibitor for AA5083 aluminum alloy in an acidic medium [3]. Expired antibacterial drugs like Ampicillin (AMP) and Ceftriaxone (CRO) served as anticorrosive agents for Sabic Fe corrosion in 1M HCl solutions [4]. Diclofenac Sodium (DSD), an expired drug, acted as a CI for Al alloy (Al6061) in acidic (0.1M HCl) and saline (3.5% NaCl) solutions [5]. An expired Salbutamol molecule was employed as a CI for mild steel (MS) corrosion in acidizing oilfield fluid [6]. Imidazole derivatives were utilized as corrosion inhibitors for steel in acidic media [7]. Levofloxacin, Moxifloxacin, Metolazone, and Nifedipine were applied as CIs for carbon steel (CS) in 2M HCl solution [8]. Telmisartan was used as a CI for Zn in 0.1M HCl medium [9]. Poly (trimethylene carbonate) (PTMC) with different molecular weights (e.g., PTMC5 and PTMC35) was applied as drug-eluting coatings on Mg alloys [10]. Ciprofloxacin (CIP) and Polymethyltrimethoxysilane (PMTMS), with an inner micro-arc oxidation (MAO) coating, were designed to enhance the corrosion resistance and antibacterial properties of Mg alloy AZ31 [11].

Ibandronate sodium (IBN) was employed as a corrosion inhibitor (CI) for a nano-coated surgical biocompatible 316L stainless steel (SS) implant in simulated body fluid (PBS) with a concentration of 10−3M at pH 7.4 and 37°C [12]. Cefadroxil (C) and Dicloxacillin (D) were utilized as CIs for aluminum (Al) in a 0.5M HNO3 solution [13]. Cefpirome and cefixime acted as CIs, self-organizing on copper (Cu) surfaces to form a bio-functional protective film. This film effectively prevents Cu corrosion during pickling and exhibits excellent resistance against Escherichia coli (E. coli) and Staphylococcus aureus (S. aureus) [14]. Urispas was employed as a CI for commercial mild steel (MS) in a 1M H_2SO_4 solution [15], while Piroxicam was used as a CI for carbon steel (CS) in a 1M HCl solution [16]. Expired Esomeprazole (20mg) served as a corrosion inhibitor for CS in desalination water (drinking water) at room temperature [17], and expired Ranitidine was employed as a CI for α-brass alloy in aqueous solution [18] and for MS in a 1M HCl solution [19]. Additionally, expired Ranitidine acted as a CI for aluminum in a 1M HCl solution [20], and expired nystatin was utilized as a CI for MS in a 0.5M HCl medium [21]. Triazepine derivatives, including 2-

amino-9- (1H-indol-3-yl) - 4- (4-methoxyphenyl)-7-oxo -1,7-dihydropyrido [1,2-b] [1,2,4] triazepine -3,8,10- tricarbonitrile (AITT) and ethyl 2-amino-8,10-dicyano-9- (2-hydroxy-3-methoxyphenyl) – 4 - (4-methoxyphenyl) -7 – oxo - 1,7-dihydropyrido [1,2-b] [1,2,4]triazepine-3-carboxylate (EHTC), were synthesized and used as CIs for MS in a 15% HCl medium [22]. Finally, expired Cefadroxil (C) and Dicloxacillin (D) were employed as CIs for aluminum in a 0.5M HNO_3 solution [23], carbohydrates have been utilized in various physical and chemical applications, including corrosion inhibition (CI), catalysts, drug delivery, coatings, etc. [24]. Natural ligands, such as carbohydrates, proteins, flavonoids, polyphenols, fatty acids, purines, pyrimidines, oleochemicals, drugs, and phytochemicals, have been investigated for their efficacy in corrosion protection and other potential applications [25]. Additionally, three new triazepine carboxylate compounds were tested as CIs for mild steel (MS) in a 1.0M HCl medium [26], while an expired drug, theophylline (TD), was used as a CI for MS in a 0.5M HCl solution [27]. Carbazodithoate was employed as a CI for carbon steel (CS) in an acidic medium [28], and expired carbimazole (CD) served as a CI for MS in a 0.5M HCl solution [29].

In the current work, the corrosion inhibition properties of expired Lactulose (La and TC) drugs for CS in a 1M HCl solution were studied using weight loss measurements and potentiometric methods.

Experimental

The carbon steel sheets had the following chemical composition (wt%): C (0.200g), Si (0.003g), Mn (0.35g), P (0.02g), and Fe (remainder). The surface area of the sample was 1.23×10^{-3} m^2. The sample was embedded in a glass tube with a diameter just larger than the sample itself. Epoxy resin (supplied by Ciba Co.) was used to adhere the sample to the glass tube. The surface of the carbon steel electrode was mechanically abraded using sandpaper, with different grades such as 1200, prior to use.

The tests were conducted using 1M HCl (supplied by Sigma-Aldrich), with the addition of various concentrations of natural product compounds. All test solutions were prepared from analytical-grade chemical reagents dissolved in distilled water and used without further purification. Freshly prepared solutions were used for each experiment. The temperature of the solutions was thermostatically controlled at the desired value, and all chemical materials had a high purity of about 99%. [30]

Apparatus

All potentiometric measurements were conducted at 25±1°C using an Orion (Model 720) pH/mV meter (Fisher Scientific). A double-junction Ag/AgCl reference electrode was employed along with a digital multimeter (TMT480012). All chemicals were of analytical reagent grade unless otherwise specified, and distilled water was used exclusively.

Testing was carried out utilizing a dielectric electrochemical cell with a volume of 250 ml. The working electrode was constructed of carbon steel (CS), with an exposed surface area to the solution of 0.00123 m^2, while the reference electrode was an Ag/AgCl electrode. Inhibitors were introduced into the test solution. The current was recorded in mA, the potential in mV, and the time in minutes.

For inhibitor measurements, the potential-versus-time dependencies of CS in the 1M HCl solution were recorded both with and without the addition of expired drugs as corrosion inhibitors (CI).

Tools and working method

A voltage and current multimeter device was utilized alongside a glass beaker with a capacity of 250 ml containing 1M HCl, titrated with Na_2CO_3 of precise concentration. An Ag/AgCl electrode served as the reference electrode, prepared by immersing two silver electrodes in 1M HCl and passing a direct current between them using a 1.5-volt battery. The working electrode was constructed of carbon steel (CS).

Cell preparation

The cell comprises two electrodes, with one being an Ag/AgCl electrode serving as the reference electrode, and the other being the (CS) electrode, with an immersed area in 1M HCl acid measuring 1.23×10^{-3} m^2. Voltage (mV) and current (mA) are recorded every ten minutes over a period of 2 hours, during which corrosion of (CS) occurs in the presence of 1M HCl acid.

Measurements of voltage, current, and time are conducted in the presence of varying concentrations of expired drugs (La. & TC) used as corrosion inhibitors (CI) at concentrations of 100, 200, 300, and 400 ppm, with each concentration tested for a duration of 2 hours.

Weight loss measurements

Experiments were performed with different concentrations of (CI). The immersion time for the weight loss is 1 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 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.

To calculate the weight loss due to corrosion using the weight loss method, you typically use the following equation:

$$W = \frac{m1 - m2}{At} ---------- (1)$$

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.

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

where W_0 and W – corrosion rate of steel in test solution without and with inhibitor, respectivelly.

All Potentiometric measurements were made at 25±1°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 silver chloride electrode, (CI) was added into the test solution. The effect of inhibition *IEe* was determined by the formula:

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

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 (CS) in the 1M HCl solution without and with the addition of (CI) were recorded.

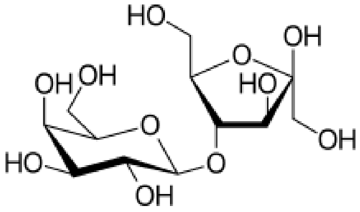
Preparation of expired drug solution

The structure, nomenclature, molar mass, & molecular formula of expired drugs one liter stock solution (1000 ppm) of (La &TC), was prepared by dissolving an accurately weighed quantity (1g) in (1L) of doubly distilled H_2O, and then the desired concentrations (100 ppm - 400 ppm) are obtained by diluting the stock solution with the desired volume of distilled H_2O. The corrosive solution, 1M HCl was prepared by diluting of high-grade HCl (42 % w, d=1. 64 g/ml, M. wt.= 36.5 g/mol.) with distilled water and dilute to up to 1M then titrated with 1M Na_2CO_3 standard solution to obtain exact 1M HCl at suitable indicator.

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

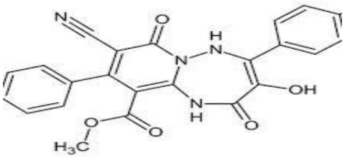
Results and discussion

Lactulose (L) (24)



4-O-β-D-Galactosyl-D-fructose.

Triazepine Carboxylate (TC)[26]



(2Cl-Me)

2H-triazepine-4-carboxylate

Chemical drugs used as corrosion inhibitors (CI) for metals or steel are among the most effective, environmentally friendly, and cost-efficient methods. This is attributed to their diverse compositions, complex structures, and large surface areas. The various functional groups present in drug molecules provide extensive coverage and protection for the surface.

The mechanism by which drug molecules act as corrosion inhibitors involves their ability to provide excellent surface coverage and protection. These molecules contain numerous electron-rich centers such as polar functional groups and aromatic rings, which facilitate their adsorption and effective action as corrosion inhibitors.

The utilization of chemical drugs as corrosion inhibitors is highly effective due to their complex structures and large surface areas. The functional groups present in drug molecules offer substantial coverage and protection for the metal surface.

In **Fig. 1**, the corrosion rate and inhibition efficiency (IEw%) for carbon steel (CS) in 1M HCl solution at 25°C are depicted both in the absence and presence of expired drugs (La & TC). It is observed that as the concentration of (La & TC) expired drugs increases, the corrosion rate decreases. In other words, the inhibition efficiency (IEw%) of (La & TC) expired drugs increases with the rise in concentration, reaching a value of 91.8%.

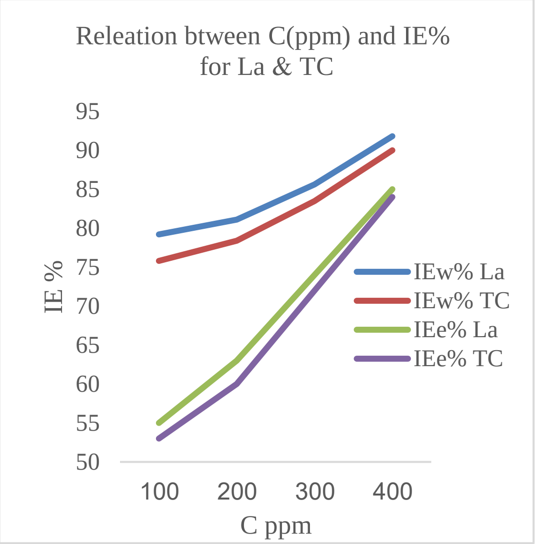


Fig.1. The effect of concentrations (in ppm) of expired drugs (La and TC) on the corrosion inhibition efficiency (IEw%) of carbon steel (CS) in a 1M HCl solution.

In Fig.1. As the concentration of inhibitors increases, the inhibition efficiency (IEw%) also increases. This is because a greater number of inhibitor molecules are adsorbed onto the surface of the carbon steel (CS), enhancing its protective effect..

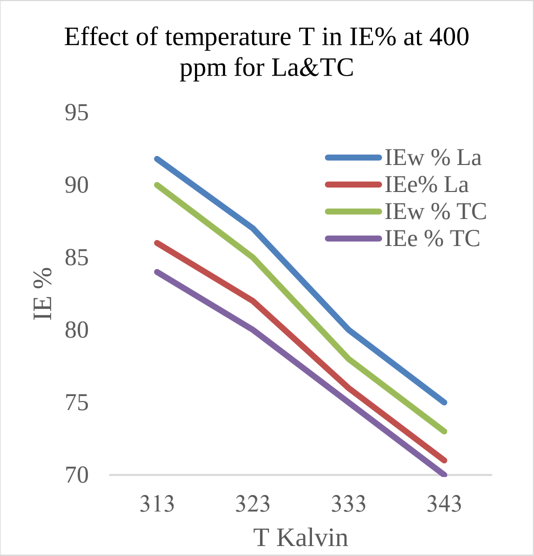


Fig.2. Effect of T (Kalvin) on IE% at 400 ppm of (La. &TC) expired drugs

In **Fig. 2**, it is demonstrated that an increase in temperature leads to a decrease in efficiency, primarily due to the heightened kinetic energy of the drug molecules adsorbed. This increase in energy results in fewer molecules being adsorbed onto the surface of the carbon steel (CS). This trend indicates that temperature primarily influences the desorption of the adsorbed inhibitor from the CS surface.

As the concentration of inhibitors increases, the inhibition efficiency (IEw%) also increases. However, when the temperature rises, the efficiency of the inhibitor decreases. This is because at higher temperatures, there is an increase in hydrogen evolution on the metal surface, leading to the desorption of the thin film formed by the inhibitor. Consequently, the inhibitor's efficiency diminishes.

Furthermore, the corrosion rate increases linearly with temperature. This observation underscores the significant impact of temperature on the desorption of the adsorbed inhibitor from the metal surface.

In Fig.3. Shows the relation between 1/*T* 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 (La &TC), Arrhenius-type equation:

$$Wcorr = Ae^{\frac{-Ea}{RT}} - - - - - (4)$$

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

In **Fig. 4.** Shows the relation between 1/*T* and Ln(*W*/*T*) for (La &TC) expired drugs, in 1M HCl to calculate, the enthalpy of activation (Δ*H**) and the entropy of activation (Δ*S**) for the corrosion of (CS) in HCl may be estimated using the transition-state equation:

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

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 −Δ*H** and from the intercepts of log(*W*/*T*)-axis, Δ*S** values were calculated.

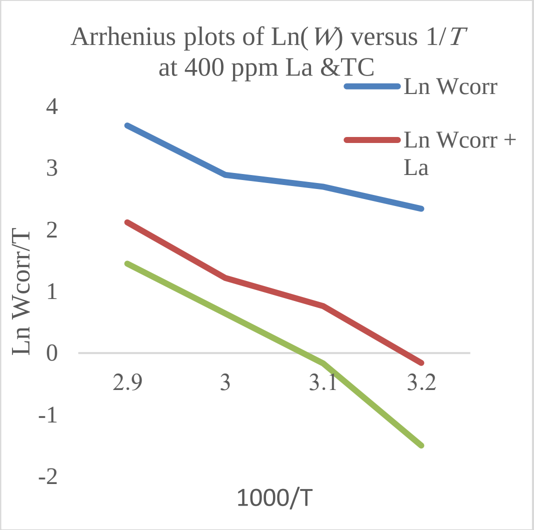


Fig. 3. Arrhenius plots of Ln (*W*) vs 1/*T* at at 400 ppm for (La &TC) expired drugs.

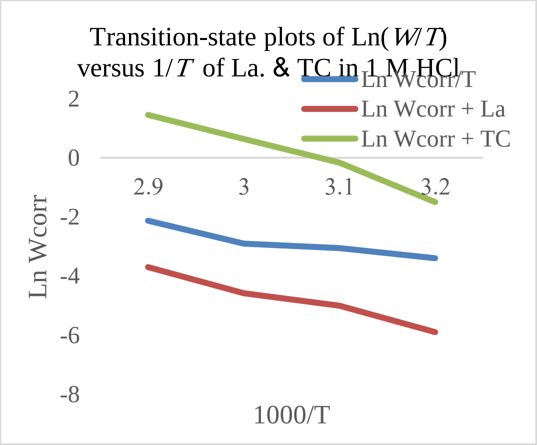


Fig.4. Transition-state plots of Ln(*W*/*T*) vs 1/*T* of (La & TC) drugs in 1M HCl in absence and presence of various concentrations of (La & TC) expired drugs.

The data was collected in **Table.1** indicate that the addition of (La & TC) leads to an increase in the activation *E*a and Δ*H** to values greater than that of the free solution. Moreover, the average difference value of the *E*a − Δ*H** is 2.6 kJ/mol which is approximately equal to the value of *RT* (2.63 kJ/mol) at the average temperature (283 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 [32].

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

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. Activation data of corrosion reaction of steel in 1M HCl in the absence and presence of expired drugs (La & TC)

	Ea (kJ/mol)	ΔH° adsorption (kJ/mol)	ΔS°adsorption (J/mol.K)
Blank	37. 4	34.8	- 35.2
La.	63.18	60.5	-60.96
TC	79.75	77.15	-79.8

The activation energies are E*a = 37. 4 kJ mol^{-1} and E*a = 63.18 kJ mol^{-1} are 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 extract 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 [33].

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. [34]

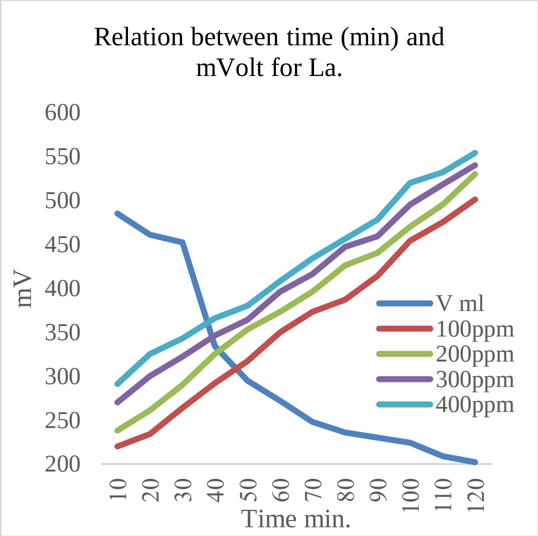


Fig. (5). Relation between time (min.) and mV at different concentration of expired (La) drug.

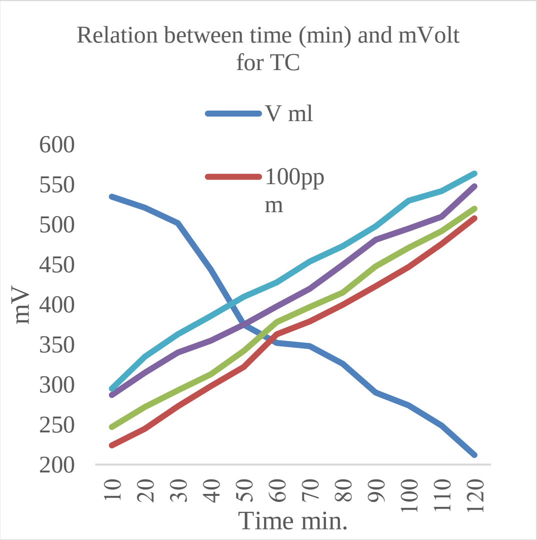


Fig.6. Relation between time (min) and mV at different concentration of expired (TC) drug.

In **Fig. 5 and 6**, the voltage behavior during the corrosion process without the addition of expired drugs (La & TC), which are used as corrosion inhibitors (CI) for carbon steel (CS) in the presence of HCl, is depicted. It is observed that the voltage decreases with the progression of the corrosion process. Over time, this decrease in voltage is attributed to the formation of an oxide layer, which enhances the conductivity of the solution. Consequently, the voltage between the working electrode and the reference electrode decreases.

However, when (La & TC) drugs are added, the voltage behavior changes. The voltage increases as a result of the adsorption of the drugs on the surface of the carbon steel (CS), leading to the formation of a protective layer between the CS and the solution. This protective layer gradually increases with time until reaching a steady state after two hours. Moreover, the voltage increases with the concentration of expired (La & TC) drugs, ranging from 100 ppm to 400 ppm.

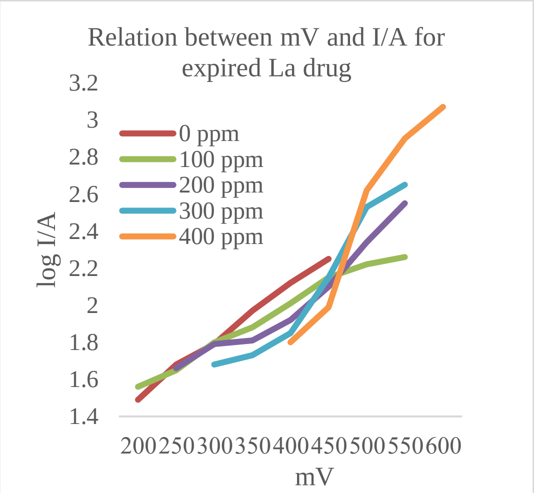


Fig.7. Relation between mV and I/A at different concentration of expired (La.) drug.

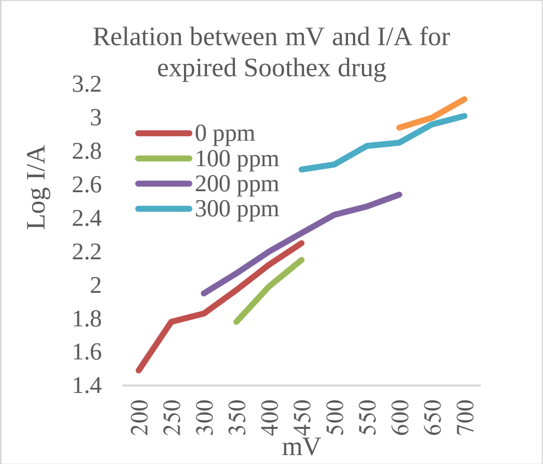


Fig. 8. Relation between mV and log I/A at different concentration of expired (TC) drug.

In **Fig. 7**, the relationship between voltage and the logarithm current density, is a measure of the electric current per unit area of the electrode surface. It is expressed in units of microamperes per square centimeter (μA/cm²) or similar. In corrosion science, the corrosion current density is directly related to the rate of the electrochemical reactions occurring at the metal surface and is a key parameter in determining the rate of metal dissolution (corrosion rate). representing the current intensity in amperes per square meter, is illustrated. It is evident that the current density is higher when the solution is without an inhibitor (0 ppm), and then decreases as the inhibitor concentration increases, holding the voltage constant. For instance, at a voltage of 400 mV, the current density decreases as the inhibitor concentration increases. However, the current generally increases with the increase in voltage.

At a concentration of 100 ppm, there is a delayed increase in voltage, and its decrease with time may be attributed to insufficient adhesion of the formed layer or the presence of multiple compounds in the drug (three compounds) with varying degrees of efficiency in the corrosion inhibition process. Nonetheless, the efficiency improves with increasing concentration up to 400 ppm.

In **Fig. 8**, it is shown that at the same voltage of 450 mV, the current density at a concentration of 100 ppm is lower than that of solutions without corrosion inhibitors (CI), but it converges with the current density at higher concentrations, particularly at 200 ppm. At higher concentrations, the voltage increases significantly, and it was not feasible to measure the current density of the inhibitor-free solution, as the cell voltage did not reach these high values except in the presence of high concentrations of the inhibitor.

Conclusion

The study revealed that the inhibition efficiency (% IEw) of the (La) drug is higher than that of the (TC) drug according to the weight loss method. However, both drugs exhibit nearly equal efficiency according to the potentiometric method. Additionally, it was observed that the (La) drug is more stable and efficient at higher temperatures, as evidenced by Arrhenius curves, transition state analysis, activation energy calculations, and changes in enthalpy and entropy.

The corrosion and inhibition processes were monitored by changes in voltage using a voltmeter and current, with a silver potential and silver chloride serving as the reference electrode. Furthermore, it was noted that the stability of the (La) drug in adsorption on the surface of carbon steel was less over time compared to the (TC) drug, rendering it more efficient in inhibiting corrosion.

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References:

[1] Wongpanya, P., Siritapetawee, J., Wongpinij, T., & Photongkam, P. (2022). Effect of antiplatelet drugs on corrosion of 316L stainless steel for application to biomaterials. *Materials Chemistry and Physics*, *278*, 125596. doi.org/10.1016/j.matchemphys.2021.125596

[2] Abdallah, M. (2002). Rhodanine azosulpha drugs as corrosion inhibitors for corrosion of 304 stainless steel in hydrochloric acid solution. *Corrosion science*, *44*(4), 717-728. doi.org/10.1016/S0010-938X(01)00100-7

[3] Chen, T., Gan, H., Chen, Z., Chen, M., & Fu, C. (2021). Eco-friendly approach to corrosion inhibition of AA5083 aluminum alloy in HCl solution by the expired Vitamin B1 drugs. *Journal of Molecular Structure*, *1244*, 130881.doi.org/10.1016/j.molstruc.2021.130881

[4] Abdallah, M., Al Bahir, A., Altass, H. M., Fawzy, A., El Guesmi, N., Al-Gorair, A. S., &Zarrouk, A. (2021). Anticorrosion and adsorption performance of expired antibacterial drugs on Sabic iron corrosion in HCl solution: Chemical, electrochemical and theoretical approach. *Journal of Molecular Liquids*, *330*, 115702. doi.org/10.1016/j.molliq.2021.115702

[5] Almashhadani, H. A., Alshujery, M. K., Khalil, M., Kadhem, M. M., & Khadom, A. A. (2021). Corrosion inhibition behavior of expired diclofenac Sodium drug for Al 6061 alloy in aqueous media: Electrochemical, morphological, and theoretical investigations. *Journal of Molecular Liquids*, *343*, 117656. doi.org/10.1016/j.molliq.2021.117656

[6] Anadebe, V. C., Nnaji, P. C., Onukwuli, O. D., Okafor, N. A., Abeng, F. E., Chukwuike, V. I., &

Barik, R. C. (2022). Multidimensional insight into the corrosion inhibition of salbutamol drug molecule on mild steel in oilfield acidizing fluid: Experimental and Computer aided modeling approach. *Journal of Molecular Liquids*, 118482. doi.org/10.1016/j.molliq.2022.118482

[7] Ouakki, M., Galai, M., & Cherkaoui, M. (2022). Imidazole derivatives as efficient and potential class of corrosion inhibitors for metals and alloys in aqueous electrolytes: A review. *Journal of Molecular Liquids*, *345*, 117815. doi.org/10.1016/j.molliq.2021.117815

[8] Abeng, F. E., Ikpi, M. E., Ushie, O. A., Anadebe, V. C., Nyong, B. E., Obeten, M. E., & Nkom, P. Y. (2021). Insight into corrosion inhibition mechanism of carbon steel in 2M HCl electrolyte by eco-friendly based pharmaceutical drugs. *Chemical Data Collections*, *34*, 100722. doi.org/10.1016/j.cdc.2021.100722

[9] Swetha, G. A., & Sachin, H. P. (2021). A study on use of telmisartan drug for corrosion inhibition of zinc in 0.1 M hydrochloric acid: surface characterization and quantum studies. *Chemical Data Collections*, *32*, 100668. doi.org/10.1016/j.cdc.2021.100668

[10] Wang, J., He, Y., Maitz, M. F., Collins, B., Xiong, K., Guo, L. & Huang, N. (2013). A surface-eroding poly (1, 3-trimethylene carbonate) coating for fully biodegradable magnesium-based stent applications: toward better biofunction, biodegradation and biocompatibility. *Acta biomaterialia*, *9*(10), 8678-8689. doi.org/10.1016/j.actbio.2013.02.041

[11] Xue, K., Liang, L. X., Cheng, S. C., Liu, H. P., Cui, L. Y., Zeng, R. C., ... & Wang, Z. L. (2021). Corrosion resistance, antibacterial activity and drug release of ciprofloxacin-loaded micro-arc oxidation/silane coating on magnesium alloy AZ31. *Progress in Organic Coatings*, *158*, 106357. doi.org/10.1016/j.porgcoat.2021.106357

[12] Fekry, A. M., & Azab, S. M. (2020). The development of an innovative nano-coating on the surgical 316 L SS implant and studying the enhancement of corrosion resistance by electrochemical methods using Ibandronate drug. *Nano-Structures & Nano-Objects*, *21*, 100411. doi.org/10.1016/j.nanoso.2019.100411

[13] Fayomi, O. S. I., Akande, I. G., Popoola, A. P. I., & Molifi, H. (2019). Potentiodynamic polarization studies of Cefadroxil and Dicloxacillin drugs on the corrosion susceptibility of aluminium AA6063 in 0.5 M nitric acid. *Journal of Materials Research and Technology*, *8*(3), 3088-3096. doi.org/10.1016/j.jmrt.2018.12.028

[14] Feng, L., Zhang, S., Tao, B., Tan, B., Xiang, B., Tian, W., & Chen, S. (2020). Two novel drugs as bio-functional inhibitors for copper performing excellent anticorrosion and antibacterial properties. *Colloids and Surfaces B: Biointerfaces*, *190*, 110898. doi.org/10.1016/j.colsurfb.2020.110898

[15] Saini, N., Kumar, R., Lgaz, H., Salghi, R., Chung, I. M., Kumar, S., & Lata, S. (2018). Minified dose of urispas drug as better corrosion constraint for soft steel in sulphuric acid solution. *Journal of Molecular Liquids*, *269*, 371-380. doi.org/10.1016/j.molliq.2018.08.070

[16] Addoun, A., Bouyegh, S., Dahmane, M., Ferroukhi, O., & Trari, M. (2019). Thermodynamic investigation on the adhesion and corrosion inhibition properties of a non-steroidal anti-inflammatory drug in HCl electrolyte applied on mild steel material. *Materials Today Communications*, *21*, 100720. doi.org/10.1016/j.mtcomm.2019.100720

[17] Gebril, A., & Bakari, I. (2022). The Inhibition Effect of the Expired Esomeprazole on the Corrosion of Carbon Steel in Desalination Water. *Journal of Pure & Applied Sciences*, *21*(1), 125-130. /doi.org/10.51984/jopas.v21i1.1419

[18] Fouda, A. S., Rashwaan, S., Ibrahim, H., & Ahmed, R. E. (2020). Corrosion Inhibition of α-brass Alloy in Aqueous Solution by Using Expired Ranitidine. *Int. J. Electrochem. Sci*, *15*, 5982-6000. doi.org/10.20964/2020.07.62

[19] Hameed, R. A. (2011). Ranitidine drugs as non-toxic corrosion inhibitors for mild steel in hydrochloric acid medium. *Port Electrochim Acta*, *29*(4), 273-285. DOI: 10.4152/pea.201104273

[20] Hameed, R. A. (2009). expired ranitidine drugs as corrosion inhibitor for corrosion of aluminium in hydrochlric acid. *Al-Azhar Bulletin of Science*, *20*(1), 151-163.Doi 10.21608/ABSB.2009.7551

[21] Mehmeti, V. (2022). Nystatin Drug as an Effective Corrosion Inhibitor for Mild Steel in Acidic Media-An Experimental and Theoretical Study. *Corrosion Science and Technology*, *21*(1), 21-31. doi.org/10.14773/cst.2022.21.1.21

[22] Paul, P. K., Mehta, R. K., Yadav, M., & Obot, I. B. (2022). Theoretical, electrochemical and computational inspection for anti-corrosion activity of triazepine derivatives on mild steel in HCl medium. *Journal of Molecular Liquids*, *348*, 118075. doi.org/10.1016/j.molliq.2021.118075

[23] Fayomi, O. S. I., Akande, I. G., Popoola, A. P. I., & Molifi, H. (2019). Potentiodynamic polarization studies of Cefadroxil and Dicloxacillin drugs on the corrosion susceptibility of aluminium AA6063 in 0.5 M nitric acid. *Journal of Materials Research and*

Technology, *8*(3), 3088-3096. doi.org/10.1016/j.jmrt.2018.12.028

[24] Yadav, M., Goel, G., Hatton, F. L., Bhagat, M., Mehta, S. K., Mishra, R. K., & Bhojak, N. (2021). A review on biomass-derived materials and their applications as corrosion inhibitors, catalysts, food and drug delivery agents. *Current Research in Green and Sustainable Chemistry*, *4*, 100153. doi.org/10.1016/j.crgsc.2021.100153

[25] Verma, C., Quraishi, M. A., & Rhee, K. Y. (2022). Natural ligands: Promising ecofriendly alternatives for corrosion protection and plethora of many prospects. *Process Safety and Environmental Protection*, *162*, 253-290. doi.org/10.1016/j.psep.2022.04.014

[26] Alaoui, K., El Kacimi, Y., Galai, M., Serrar, H., Touir, R., Kaya, S. A. V. A. Ş., ... & Ebn Touhami, M. (2020). New triazepine carboxylate derivatives: correlation between corrosion inhibition property and chemical structure. *International Journal of Industrial Chemistry*, *11*, 23-42. doi.org/10.1007/s40090-019-00199-5

[27] Khalifa, S., AL-abbassi, A., & Suliman, M. (2018). Adsorption and Corrosion Inhibition of Mild Steel in Acidic Media by Expired Pharmaceutical Drug. *Journal of Pure & Applied Sciences*, *17*(4). doi.org/10.51984/jopas.v17i4.414

[28] Suliman, M., Alkilani, M., Abdullah, T., & Ali, M. (2019). The Effect of Methyle Carbazodithoate" Corrosion inhibitor on Corrosion Behaviour of Low Carbon Steels in Acid Solution at Different Temperatures. *Journal of Pure & Applied Sciences*, *18*(4). doi.org/10.51984/jopas.v18i4.426

[29] Al-abbasi, A., & Shana, I. E. (2021). Corrosion Inhibition of Mild Steel in Acidic Media by Expired Carbimazole Drug. *Journal of Pure & Applied Sciences*, *20*(2), 176-180. doi.org/10.51984/jopas.v20i2.1646

[30] Matter A.B. Ayad, T. M. (2022). Study The Effect of Extract berry and mango leaves as a corrosion inhibitor of Cu, Carbon steel and Oil Pipelines. *Journal of Pure & Applied Sciences*, *21*(1), 154-164. doi.org/10.51984/jopas.v21i1.1671

[31] Verma, C., Quraishi, M. A., & Rhee, K. Y. (2021). Present and emerging trends in using pharmaceutically active compounds as aqueous phase corrosion inhibitors. *Journal of Molecular Liquids*, *328*, 115395. doi.org/10.1016/j.molliq.2021.115395

[32] El Ouali, I., Hammouti, B., Aouniti, A., Ramli, Y., Azougagh, M., Essassi, E. M., & Bouachrine, M. (2010). Thermodynamic characterisation of steel corrosion in HCl in the presence of 2-phenylthieno (3, 2-b) quinoxaline. *J. Mater. Environ. Sci*, *1*(1), 1-8. www.jmaterenvironsci.com

[33] Hamdy, A., & El-Gendy, N. S. (2013). Thermodynamic, adsorption and electrochemical studies for corrosion inhibition of carbon steel by henna extract in acid medium. *Egyptian Journal of Petroleum*, *22*(1), 17-25. doi.org/10.1016/j.ejpe.2012.06.002

[34] Traxler, I., Singewald, T. D., Schimo-Aichhorn, G., Hild, S., & Valtiner, M. (2022). Scanning electrochemical microscopy methods (SECM) and ion-selective microelectrodes for corrosion studies. *Corrosion Reviews*, *40*(6), 515-542. doi.org/10.1515/corrrev-2021-0104.