Carbon steel corrosion inhibition in acidic medium by expired drugs

تثبيط تأكل الفولاذ الكربونى فى الوسط الحامضى باستخدام الادوية منتهية الصلاحية

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Abstract

Corrosion and expired pharmaceutical materials represent a research spot because of their important issues in earth planet and human life. Three different expired drugs were bought from pharmacies in Baghdad city, Iraq and tested to determine their efficiencies in solving carbon steel corrosion in 1M HCl solution.

Various exposure times were subjected to test Citicoline (Samozina, syrup, 100 mg in 1mL); Carbocisteine (5%) (Rhinathiol syrup, 750 mg); or Paracetamol (Panalife syrup 120 mg in 5mL) as an inhibitor protect carbon steel specimen from being corroded. Inhibition efficiencies (IE%) of the tested drugs were ranged from low to high values reflecting the effects of drug- active component type, time, concentration, and acidic hydrolysis as a suggested explanation.

Temkin isothermal model was applied and K_{ads} were ranged from [(26.43374-183.549), (10.36883 -311.2802), and (3.499144 - 61.50259)] for Citicoline, Carbocisteine, and Paracetamol respectively. While Δ G_{ads} values (in negative sign) were varied from (18.0494-22.8559, 15.7418-24.1738, and 13.033-20.1562) for Citicoline, Carbocisteine, and Paracetamol respectively. From Δ G_{ads}values, it can be concluded that physisorpion than chemisorption was occurred.

Keywords: carbon steel, expired drugs, hydrochloric acid, corrosion.

الخلاصة

يمثل التأكل والمواد الصيدلانية منتهية الصلاحية بقعة البحث مهمة بسبب اهميتهما لكوكب الارض والحياة البشرية. تم شراء ثلاث ادوية منتهية الصلاحية من الصيدليات في مدينة بغداد – العراق وفحصها لتحديد كفاءتها في ايجاد حل لمسألة تأكل الفولاذ الكربوني في محلول حامض الهيدر وكلوريك 1M.

تم تطبيق اوقات مختلفة لاختبار

Citicoline (Samozina, syrup, 100 mg in 1mL); Carbocisteine(5%) (Rhinathiol syrup, 750 mg);

او (Panalife syrup 120 mg in 5mL) في حماية عينة الفولاذ الكربوني من التأكل. تراوحت كفاءة التثبيط (IE)) للادوية تحت الفحص من القيم القليلة الى العالية كانعكاس تأثيرات كل من نوع الدواء- المكون الفعال من الأدوية تحت الإختبار من الأقل إلى القيم العالية التي تعكس آثار نوع المادة الفعالة ضمن الدواء، والوقت، والتركيز، والتحلل الحمضية كتفسير مقترح.

تم تطبيق Temkin نموذج متساوي الحرارة وتراوحت K_{ads} من [(183،549-26،43374)، (183،56883)، (10،36883)، (10،36883)، (20،311،2802)، (20،311،2802)، (20,311،2802)، (20,311،2802)، (20,311،2802)، (20,311,2802). (20,311,2802

في حين تفاوتت قيم G_{ads} لمَّ(إشارة سالبة) من (18،0494-22،8559) 24،1738-24،1738-24،13،033 و13،033-10 Paracetamolو Paracetamol او Citicolinعلى التوالي. من القيم G_{ads} م فإنه يمكن استنتاج أن الامتزاز الفيزياوي كان حاصلا اكثر من الامتزاز الكيمياوي.

الكلمات المفتاحية: الفولاذ الكربوني ، الادوية منتهية الصلاحية ، حامض الهيدر وكلوريك ، تأكل.

Introduction

Among many serious problems that concern human life, there are two important issues affected living organisms on earth which are corrosion and pollution. The first problem depends on its basic thermodynamic reaction or process companied with decreasing in Gibb's free energy [Bockris and Reddy 2002].

It is known that corrosion results affects industrial applications and processing especially in high cost protection methods or maintenance of metallic or non-metallic materials. To solve metallic corrosion, various solution had been applied around the world with many advantages and disadvantages including: coatings (biofilm painting, enamel, and plating), (organic, inorganic, organo-metallic) inhibitors, anodization, and cathodic protection [Fontana and Greene, 1987; Bard and Faulkner, 2001, Ahmed, 2006, Lece et al., 2008; Şahin et al., 2008; Al-Obaidi, 2013; Al-Obaidi, 2015; Kubba et al., 2016].

The other problem that took place of scientific attentions was the management of expired drugs because of their health effects on environment. Drugs may containing several heteroatoms in their backbone such as N, O, S, P that are important factor in choosing any corrosion inhibitor with organic base [Newman and Cragg, 2007].

The main important limiting of any corrosion inhibitor success is its eco-friendly situation beside its inhibition efficiency. So, the use of drug as anti-rusting agent depends on its positive effect on environment especially aquatic, natural origin, cost, presence of polar or non-polar, hydrophilic, lipophilic, biodegradable, or persistency character. [Raja and Sethuraman, 2008]

Eliminating of many synthesis steps of new organic derivative that may be included usage of toxic or may be defined as carcinogenic materials (starting materials, catalyst, solvent, ...) can be also putted as essential issues in solving corrosion techniques. Also, drugs are well identified materials by all needed characteristic techniquesbefore applying as corrosion inhibitors.

Many scientific published papers demonstrated the use of drugs as good corrosion inhibitors against different conditional environments. These publication may be categorized to β -lactam[Eddy et al., 2009; Eddy and Odoemelam, 2008; Gece, 2011], quinolone, tetracyclines, macrolide, lincosamide, sulphonamide, aminoglycoside, amphenicol, antifungal, amoebicidal, antiviral, antihypertensive, antipsychotic, anthelmintic, muscle relaxant, opioid analgesic, and histamine classes.

The other direction of corrosion research subject was taken advantage of expired drugs as effective inhibitors to this important industrial problem. They present a promising choice that decreasing needed awareness of environmental protection.

Due to the above scientific, economic, environmental, and industrial considerations, our main goal was directed to use several expired drugs as anti-rusting agents purchased from pharmacies located in Baghdad city, Iraq with different traditional marks. This goal also was planned to be achieved

with weight loss method for carbon steel in acidic media and made use of Temkin isotherm as an adsorption mechanism model.

Experimental section

Instruments:

SpectroMax_x, stationary metal analyser, AMETEK Spectro-Analytical Instrument (Germany, 2012 / model), in State Company for Inspection and Engineering Rehabilitation (S.I.E.R), Ministry of Industry and Minerals was applied for elemental analysis of carbon steel specimen.

Chemicals

All required materials: acid (hydrochloric), ethanol, and acetone were purchased from trusted international companies (BDH, Fluka, Merck). The expired drugs under investigation were obtained from pharmacy shops in Baghdad city, Iraq and their details as follows:

1. Citicoline:

Trade name: Samozina, syrup; contains: 100 mg in 1mL; source: Ferrer International, Spain; Manufacturing date: 7/2012; Expired date: 7/2015; experiment date: Jan. 2016.

2. Carbocisteine (5%):

Trade name: Rhinathiol syrup; contains: 750 mg; source: Unither Liquid, France; Manufacturing date: 8/2012; Expired date: 8/2015; experiment date: late of March. 2016.

3. Paracetamol

Trade name: Panalife syrup; contains: each reaspanful 5mL contain paracetamol 120 mg; source: LIFE Pharma FZE, Jebel Ali, Dubai, UAE; Manufacturing date: 3/2013; Expired date: 3/2015; experimentdate: middle of February, 2016.

Weight loss measurements in acidic medium [Al-Obaidi, 2013; Al-Azzawiand Hammud, 2014] Specimen Characterization: The material composition was characterized with SpectroMax_x/ stationary metal analyser and its actual metallurgical type (in percent) which were Carbon:0.187, Silicon: 0.311, Manganese:1.03, Phosphor:0.007, Sulfur: 0.012, Chromium: 0.003, Molybdenum : 0.002, Nickel: 0.029, Aluminum : 0.038, Cobalt: 0.001, Copper: 0.002, Tungsten : 0.222, Arsenic: 0.0083, Iron, 98.3 beside V, Pb, Sn, Zr, Bi, Ca, Ce, B, Zn, and La in trace amounts.

The carbon steel sheets (2 cm x 2 cm x 0.02 cm) abraded with emery paper [(320-500-800) grade]were washed with distilled water and acetone. After weighing accurately, the specimens were immersed at inclined position in beakers which contained 25 mL of the studied hydrochloric acid (1M) with different concentrations of the inhibitor.

All the aggressive solutions (with or without expired drug presence) were open to air. The specimens were taken out after a specific time, washed, dried, and weighed with accuracy.

Concentration	IE%					
	1 hr.	2hrs.	4 hrs.	24 hrs.		
0.5 mL	34.5	37	16			
0.004 M	54.5	57	10	-		
1 mL	49	59	42	28		
0.00784 M	47	57	+2	20		
1.5 mL	47	52.4	38.7	25		
0.0113 M	+/	52.4	50.7	23		

Table -1-: Inhibition efficiency (IE%) of carbon steel with Citicoline as expired drug in 1M HCl.

Table -2-: Inhibition efficiency (IE%) of carbon steel with Carbocisteine (5%) as expired drug in 1M HCl.

Concentration	IE%					
	1 hr.	2hrs.	4 hrs.	24 hrs.		
0.5 mL 0.00547 M	55	63	61	54		
1 mL 0.0107 M	33	38	25	-		
1.5 mL 0.0157 M	44	47	27	14		
2 mL 0.0207 M	66	72	71	65		
3 mL 0.0298 M	77	77	82	75		

Table -3-: Inhibition efficiency (IE%) of carbon steel with Paracetamol as expired drug in 1M $\,$

HCl.							
Concentration	IE%						
Concentration	1 hr.	2hrs.	4 hrs.	24 hrs.			
0.5 mL 0.00312 M	72	71	60	83			
1 mL 0.0061 M	87	73	50	77			
1.5 mL 0.00898 M	88	76	61.6	80			
2 mL 0.0117 M	91	85	68.7	87			
3 mL 0.01701 M	84	73	62.5	88			

Concentration	1 h	ır.	2hrs.		4 h	rs.	24 hrs.	
	K _{ads}	- ΔG	K _{ads}	-ΔG	K _{ads}	-ΔG	K _{ads}	-ΔG
0.5 mL	131.6794	22.0423	146.8254	22.3121	47.61905	19.5223	-	-
1 mL	122.549	21.8555	183.549	22.8559	92.36453	21.1552	49.60317	19.6155
1.5 mL	26.43374	18.0494	97.4195	21.2785	55.86915	19.9021	29.49853	18.321

Table -4-: Temkin isothermal model results (K_{ads}) and Gibbs free energy (ΔG , KJ/mol.)of carbon steel with Citicoline as expired drug in 1M HCl.

Table -5-: Temkin isothermal model results (K_{ads}) and Gibbs free energy (ΔG, KJ/mol.)of carbon steel with Carbocisteine (5%) as expired drug in 1M HCl.

Concentration	1 hr.		2hrs.		4 hrs.		24 hrs.	
	K _{ads}	- ΔG	K _{ads}	- ΔG	Kads	- ΔG	K _{ads}	- ΔG
0.5 mL	223.441	23.3524	311.2802	24.1738	285.942	23.9635	214.6093	23.2471
1 mL	46.03152	19.4304	57.28068	19.9719	31.15265	18.4635	-	-
1.5 mL	50.0455	19.6296	56.4836	19.9292	23.55815	17.7643	10.36883	15.7418
2 mL	94.23187	21.1876	124.8266	21.8834	118.8483	21.7619	90.15257	21.0988
3 mL	112.3432	21.6139	112.3432	21.6139	152.871	22.3759	100.6711	21.3722

Table -6-: Temkin isothermal model results (Kads) and Gibbs free energy (ΔG, KJ/mol.) of carbon steel with Paracetamol as expired drug in 1M HCl.

Concentratio	1 h	ır.	2hrs.		4 hrs.		24 hrs.	
n	K _{ads}	- ΔG	Kads	- ΔG	Kads	- ΔG	K _{ads}	- ΔG
0.5 mL	32.3920	18.567	30.8407	18.446	18.7387	17.211	61.5025	20.156
0.3 IIIL	7	6	3	1		6	9	2
1 mL	42.1512	19.212	17.0291	16.967	5.81396	14.306	21.0861	17.497
1 mL	2	4	7	8	2	3	4	
1.5 mI	30.7924	18.427	13.2967	16.348	6.73585	14.664	16.7958	16.926
1.5 mL	6	3	5	4	1	8	9	8
2 mL	31.8422	18.502	17.8456	17.069	6.69211	14.642	21.0756	17.481
2 mL	1	8	3	9	2	8	1	6
3 mL	10.2504	15.691	5.67638	14.229	3.49914	13.033	15.3962	16.697
	3	6	8	7	4	15.055	3	8

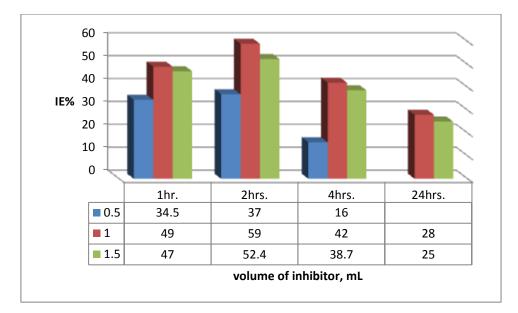


Figure -1-: Inhibition efficiency (%) of Citicoline as expired drug in 1M HCl.

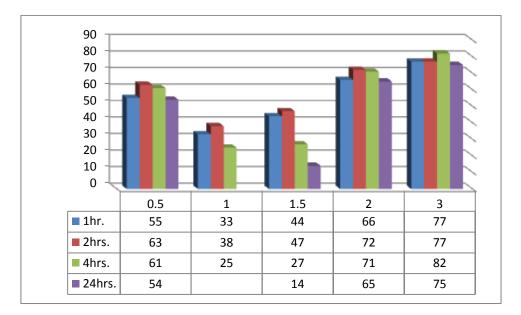


Figure -2-:Inhibition efficiency (%) of Carbocisteine (5%) as expired drug in 1M HCl.

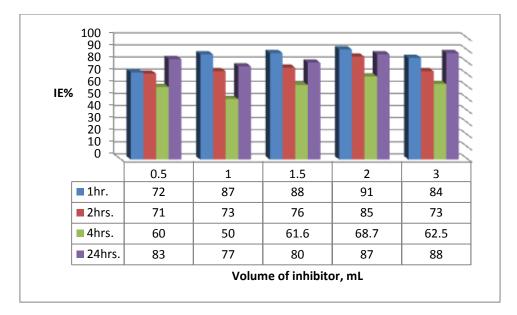


Figure -3-:Inhibition efficiency (%) of Paracetamolas expired drug in 1M HCl.

Results and Discussion

Nowadays, many important issues faced human on the earth planet especially in safety and environment including corrosion and expired or unused materials. These problems can be a solution to each other's by applying expired drugs as corrosion inhibitors for different metals or alloys in various corrosive conditions (medium, temperature, time,...). With this solve, many industrial and technological applications can be a free of damage that resulted from rusting. It is not new for such solution in research area where scientific publications demonstrated and reviewed these ideas [Vaszilcsin et al, 2012; Abdel Hameed et al., 2015].

Our research group focused on three expired drugs bought from Iraqi pharmacies located in Baghdad city where the detailed information presented in previous mentioned section: experimental- chemicals. From tables (1-3) and Figures (1-3), it can be noticed several important points as bellows:

Samozina drug with its main effective component (Citicoline) was tested with different volumes (0.5, 1, and 1.5) mL that added to 1M HCl solution containing carbon steel specimen with exposure time ranged from 1 hour to 24 hours.

- a. IE% values were ranged from 16% to 59% in general.
- b. In spite of containing Citicoline(Figure -4-) different moieties with heteroatoms (N, O, P) such as nucleosidicCytidinyl; with its primary amino group; and phosphatidylcholine, thesemoieties did not increased inhibition efficiency% (IE%) to high degree.
- c. Also, exposure time played important role in decreasing IE% values which might be related to the action of linked phosphate anion formed by the action of strong acidic medium (HCl, 1M) and long hydrolysis time (Equation -1-).

This explanation can be proved with further hydrolysis experiments which were not done in this short study. Furthermore, the hydrolysis of this drug is known to be achieved in intestine to cytidineand choline [Wurtmanet al., 2000].

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d. The moderate to low IE% values can be subjected to low concentrations [(0.004, 0.0078, 0.0113) mole/liter] of the tested expired drug where it contained 100 mg in 1mL as an active component.

According to our readings, this is the first time that this drug or active components was putted in test with corrosion issue. Also, we did not find any scientific information that can be used in explanation of this action in such medium.

Bindaiya and Argal confirmed with their published research article in 2013 that Citicoline Monosodium Tablet (500mg) formulation was stable under tested conditions that confirmed with the usage of HPLC instrumentation. The tested conditions were acidic: (0.01, 0.1, 1, 2, 5) N of HCl , basic: (0.01, 0.1, 1, 2, 5) N of NaOH , neutral: water, oxidative: (1, 3, 10, and 30) % of H₂O₂ v/v, thermal: (40, 60, 80, and 100)⁰C and time period (2, 4, 8, 12, and 24) hours in hot air oven, and photolytic: sunlight for 1day, 2 days, 3days and 7days [Bindaiya and Argal, 2013].

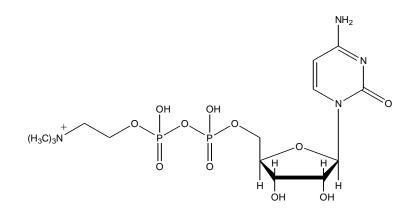
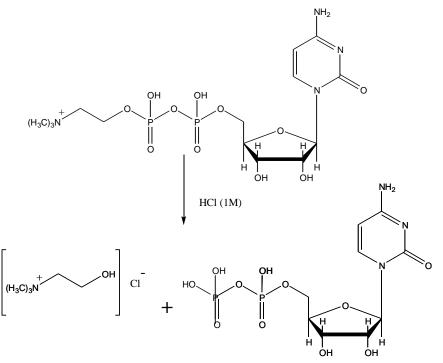


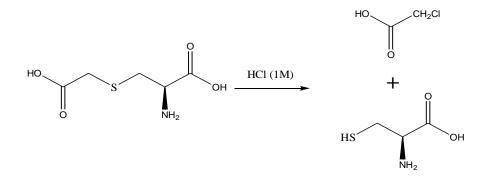
Figure -4-: Chemical structure of Citicoline.



Equation -1-: Suggested hydrolysis mechanism of Citicoline in strong acidic medium (HCl, 1M).

The second tested drug was Carbocisteine (5%) syrup that it is known with many trade names including Rhinathiol containing 750 mg of alkylated cysteine as an active component.

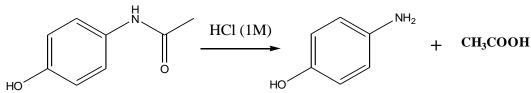
- a. Its IE% values were ranged from 14% to 82% (Table -2-, and Figure -2-).
- b. From Table -2- and Figure -2-, it is clear that applying more added volumes than the previous discussed drug (Citicoline) had a great influence on IE% values.
- c. Presence of (N, O, S) as heteroatoms in the chemical structure of this drug might be directed IE% to high values with Carbocisteine than Citicoline.
- d. The wide range in IE% results may be attributed to the effect of strong acidic medium (1M, HCl) on tested carbon steel with presence of small quantities of expired drug that may also hydrolyzed to their essential components that formed from as suggested below (Equation -2-):



Equation -2-: Suggested hydrolysis mechanism of Carbocisteine in strong acidic medium (HCl, 1M).

The third expired drug that applied as a corrosion inhibitor was paracetamol which showed more effectiveness in inhibition than both previous mentioned drugs. The IE% range of paracetamol was from 50% to 91% for five added volumes at different period of exposure time (1, 2, 4, and 24) hours. The increasing in IE% values of this drug may be related to its different characters as below(Table -3- and Figure -3-):

- a. This drug contains acetoaminophen as an active component with N and O heteroatoms attached to aromatic phenyl ring.
- b. It is known that paracetamol can be hydrolyzed to 4-aminophenol in HCl solution as amide hydrolysis (Equation -3-). This note may explain the increasing in IE% compared with other two expired drugs.



Equation -3-: Suggested hydrolysis mechanism of Paracetamol in strong acidic medium (HCl, 1M).

The adsorption Mechanism:

To study the adsorption mechanism, several important factors had a great influence on it such as time, temperature, chemical and physical properties of the tested metal or alloy, corrosive medium, and inhibitor. These factors are effective in corrosion processing and inhibition of it especially chemisorption.

It is known that chemisorption involves sharing or transferring of the negative charge from inhibitor to the metal or alloy surface by forming coordination bonding through the presence of heteroatoms (N, O, S, ..) [Ahamad et al, 2010; Yadav and Quraishi, 2012; Al-Azzawi and Hammud, 2014; Kubba et al, 2016].

In the same subject area, adsorption isotherm [Quraishi et al., 2000] represented a linear relationship between coverage area (θ) and the inhibitor concentration (C_{inh}) as in Temkin [K_{ads} C = $e^{-f\theta}$], Frumkin [K_{ads} C = ($\theta/1-\theta$) $e^{-f\theta}$] or Langmuir isotherm [K_{ads} C = $\theta/1-\theta$] where f (θ) depends upon the physical model and K_{ads} is the equilibrium constant of adsorption.

In this study, the average values for free energy of adsorption (ΔG_{ads}), were calculated by using the following equation (Temkin isotherm) (Tables:-4- to -6-):

$$K = \theta / (1 - \theta) C$$

$$K = (1/55.5) \exp (\Delta G_{ads} / RT)$$

where θ is degree of coverage on metal surface, C is concentration of inhibitors in mol/L, R is molar gas constant in J/K. mol and T is temperature. The value of 55.5 is water concentration (M).

Generally, $\Delta G^{\circ}_{ads} \leq -20 \text{ kJ/mol signifies physisorption with Coulombic electrostatic interactions exist between the charged molecules and the charged metal surface while more negative than -40 kJ/mol for chemisorption and these negative values clarifies spontaneous adsorption [Quraishi et al., 2000] with formation of the chemical bond between the inhibitor and the metal surface. Also, higher values of K_{ads} refer to higher adsorption and inhibiting effect [Saliyan and Adhikari, 2008].$

Our finding of K_{ads} were ranged from (26.43374, 10.36883, and 3.499144) to (183.549, 311.2802, and 61.50259) for Citicoline, Carbocisteine, and Paracetamol respectively. While Δ G_{ads}values (in negative sign) were varied from (18.0494, 15.7418, and 13.033) to (22.8559, 24.1738, and 20.1562) for Citicoline,Carbocisteine, and Paracetamol respectively. From Δ G_{ads}values, it can be concluded that physisorpion than chemisorption.

Conclusion:

The physisorpion of three expired drugs under test (Citicoline, Carbocisteine, or Paracetamol) with weight loss method on carbon steel – HCl solution was the concluded from Temkin Model calculations (ΔG°_{ads}). These obtained results explained the moderation in corrosion inhibition values that may be adjusted with hydrolysis expectation in high acidic medium (HCl, 1M). This work is a good a tempt to solve both corrosion and expired drug problems.

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