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RESEARCH ARTICLE **CORROSION**

Investigation of Expired Esomeprazole's Effectiveness as a Corrosion Inhibitor for Brass in Gabraun Water

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ABSTRACT

This study aimed to investigate the effectiveness of Esomeprazole as an inhibitor for Brass corrosion in Gabraun water. Brass is an ideal alloy for transporting water through pipes and fittings and is also suitable for use in marine engines and pump parts. The tests were conducted at room temperature, using Gabraun water as the electrolyte. The weight loss method was employed, and both open and closed systems were used for the testing. Results indicated that the weights of all specimens, both with and without the inhibitor, decreased after the tests. The lowest corrosion rate recorded was 0.04139 cm/year for a specimen with an area of 8.545 cm² and an inhibitor amount of 1.5g. Importantly, the color of the Gabraun water did not change, which is attributed to the effect of the inhibitor. However, there was no significant difference in corrosion rates between the specimens tested with the inhibitor and those without it. This suggests that while Esomeprazole is not effective in reducing the corrosion rate of Brass, it does inhibit the reaction between copper (Cu) and sulfate (SO_4). Additionally, Gabraun water proved to be a corrosive medium when used as an electrolyte for Brass testing, which contrasts with the findings of the Langelier Saturation Index (LSI).

دراسة فعالية الإيزوميبرازول منتهي الصلاحية كمثبط للتآكل على سبيكة النحاس الأصفر في مياه قبرعون

صلاح الدين سالم التحبو [1]، *، إبراهيم بكاري العربي [1]، عبد السلام عبد الله جبريل [1]

الملخص

الهدف من هذه الدراسة كان التحقيق في فعالية الإيزوميبرازول كمثبط لتآكل البراص في مياه قبرعون. يعتبر البراص سبيكة مثالية لنقل المياه عبر الأنابيب والتجهيزات، كما أنه مناسب للاستخدام في المحركات البحرية وأجزاء المضخات. تم إجراء الاختبارات في درجة حرارة الغرفة باستخدام مياه قبرعون كإلكتروليت. تم استخدام طريقة فقدان الوزن، وتم استخدام أنظمة مفتوحة ومغلقة في الاختبارات. أظهرت النتائج أن وزن جميع العينات، سواء مع المثبط أو بدونه، قد انخفض بعد الاختبارات. أقل معدل تآكل تم تسجيله كان 0.04139 سم/سنة لعينة بمساحة 8.545 سم² وكمية من المثبط تبلغ 1.5 جرام. من المهم أن لون مياه قبرعون لم يتغير، ويعزى ذلك إلى تأثير المثبط. ومع ذلك، لم يكن هناك فرق كبير في معدلات التآكل بين العينات التي تم اختبارها مع المثبط وتلك التي تم اختبارها بدونه. وهذا يشير إلى أنه بينما لا يكون الإيزوميبرازول فعالًا في تقليل معدل تآكل البراص، إلا أنه يمنع التفاعل بين النحاس (Cu) والكبريتات (SO_4). بالإضافة إلى ذلك، تبين أن مياه قبرعون هي وسط تآكل عند استخدامها كإلكتروليت لاختبار البرونز، وهو ما يتناقض مع نتائج مؤشر التشبع لانجيليه (LSI).

الكلمات المفتاحية

بحيرة قبرعون
سبيكة البراص
معدل التاكل
ايزوميبرازول
مثبطات التاكل

Introduction

Gabraun Lake, located in southern Libya, is a saline lake situated in the Libyan Desert. Its water exhibits a thermal gradient, with a cooler surface (approximately 7.5 meters deep) that becomes hotter at greater depths [1]. A substantial body of research has examined the environmental aspects of the lake[2–4]; however, further investigation and engineering studies are necessary to gain a deeper understanding of its dynamics and to address the complex challenges it presents. A significant body of research has investigated the use of plants and antioxidants as environmentally friendly corrosion inhibitors[5,6]. However, a notable gap remains in the literature regarding the potential of expired drugs as corrosion inhibitors. Further exploration in this area is warranted to assess the viability and efficacy of expired pharmaceuticals in corrosion mitigation, particularly in comparison to more widely studied alternatives. Globally, the disposal of unused or expired medications as household waste is widespread, affecting nations of all income levels. [7], [8]. Studies show that fish and vegetables are contaminated with heavy metals, linked to human activities and improper waste disposal in terrestrial and marine environments[9], [10]. Kudari et al. [11] studied the fate of medicines after expiry. Their findings clearly indicated that expired drugs retain stability, quality, and efficacy. Esomeprazole ($C_{17}H_{19}N_3O_3S$), marketed as Nexium, is a medication that reduces stomach acid by inhibiting the

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H+/K+-ATPase enzyme in parietal cells. It is used to treat gastroesophageal reflux disease and peptic ulcer disease [12]. Like other proton pump inhibitors, its efficacy stems from this mechanism of action. Corrosion refers to the chemical or electrochemical attack caused by the surrounding environment [13]. Brass, an alloy of copper and zinc, generally resists atmospheric corrosion well. The proportion of zinc influences the alloy's properties, including its color, which can range from red to yellow [14]. It is an ideal alloy for transporting water through pipes and fittings and is also suitable for use in marine engines and pump parts. Previous studies support the use of chemical inhibitors for copper alloys[15]. Nasser et al.:[16] demonstrated that organic compounds containing sulfur groups, such as diethyl sulfide, effectively inhibit copper corrosion in nitric acid. Similarly, El-Sherbini et al.[17] found that sodium molybdate retards copper dissolution in hydrochloric acid. The electrochemical behavior of copper is illustrated by its Pourbaix diagram (Figure 1) [18]. Copper, which is more stable than iron in water, undergoes corrosion in acidic and highly alkaline electrolytes under high oxidizing potentials[18].

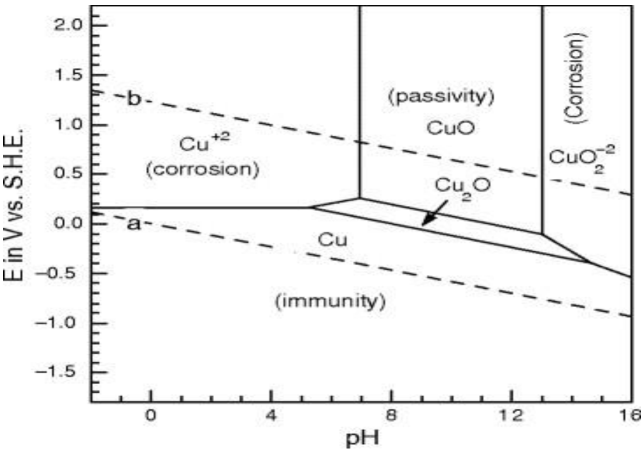

Fig. 1: Pourbaix diagram for the copper–water system at25◦C[18]

According to Gebril et al. [19], the Gabraun water utilized in brass corrosion tests exhibited a color change to dark blue, which signifies a chemical reaction between copper and sulfate, resulting in the formation of copper sulfate. The Esomeprazole is a medication can reduce stomach acid for the humankind and the effectiveness is similar to other proton pump inhibitors, this study aims to evaluate esomeprazole's efficacy as a corrosion inhibitor for brass in Gaberoun Lake's saline water, while exploring the repurposing of pharmaceutical waste for environmental applications.

Methods and Materials

In this study, brass, an alloy of copper, was utilized, and its composition was analyzed through X-ray diffraction at the Faculty of Science, Sebha University, as detailed in Table 1.

Sixteen (16) specimens were cut from Brass rod, twelve with radius of 10mm and 20mm long and four with radius of 10mm and 9mm long as shown in figure (2). All specimens were polished with 400 and 1200 grit emery paper to eliminate oxides and contaminants from their surfaces.

The chemical analysis of the Gabraun water used in this study, conducted at Sebha University's Faculty of Science, is shown in Table 2.

Corrosion Tests

The test solutions in this research consisted of Gabraun water samples. The experiments were conducted in 600 ml beakers, and different quantities of expired Esomeprazole inhibitor ($C_{17}H_{19}N_3O_3S$) were incorporated with the water in the beakers.[20]. Esomeprazole was mixed into Gabraun water using an electric stirrer for a few minutes until the esomeprazole dissolved. Each specimen was completely immersed in its respective beaker. Prior to and following the corrosion tests, all specimens were thoroughly cleaned and weighed. These tests were carried out at a room temperature of 25°C over a period of 49 days.

Table 1: Chemical Composition of the Brass

Element	Masses %
Cu	57.16
Zn	36.69
Si	1.19
Pb	2.47
Al	1.54
Fe	0.3313
Ni	0.0266
Sn	0.264
Co	0.0082
Mo	0.0112
Au	0.0206
Pt	0.0293
Cd	0.0029
Ca	0.023

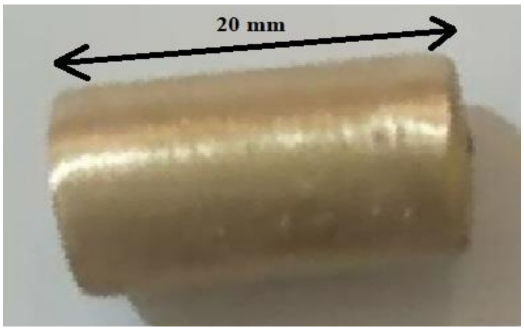

Fig. 2: Brass specimen with radius 10mm and 20 mm long before immersion

Table 2: Analysis of Gabraun's water

Parameter	Gabraun water before immersion
Ph	9.66
Conductivity (µS/cm)	162500
Salinity (ppt)	27
TDS (mg/L)	24780
Total Alkalinity(mg/L)	430
Hardness Ca^{++} (mg/L)	83
Hardness Mg+2(mg/L)	28
SO4 (mg/L)	37
K+ (mg/L)	34
Na+(mg/L)	135
Cl- (mg/L)	95
Total Hardness (mg/L)	384

Langelier Saturation Index (LSI)

To predict how Gabraun water reacts with calcium carbonate, the Langelier Saturation Index (LSI) was developed to assess calcite formation and is used as an indicator of corrosion. Additionally, it serves as a key factor in driving scale formation and growth, with pH being the primary influencing variable[21]. The alkalinity (mg/L, as $CaCO_3$, or calcite), the actual pH, the temperature of water (°C), the calcium hardness (mg/L Ca^{+2} as calcium carbonate), and the total

dissolved solids (TDS) (mg/L TDS) are the important parameters to calculate LSI[21].
LSI is defined as the following equation [21]:

$$LSI = pH - pHs \quad (1)$$

Where:
pH is the measured water pH.
pHs is the pH at saturation in calcite or calcium carbonate and is defined as:

$$pHs=(9.3+A+B)-(C+D) \quad (2)$$

$A=\{\log_{10}[TDS]-1\}/10$
$B=-13.12\times\log_{10}(^0C+273)+(34.55)$
$C=\log_{10}[Ca^{2+} \text{ as } CaCO_3]-[0.4]$
$D=\log_{10}[\text{alkalinity as } CaCO_3]$

The LSI indicates by the following [22]:
The negative LSI: The water will dissolve $CaCO_3$, in this case no potential to scale.
The positive LSI: this indicates that Scale can form and the precipitation of $CaCO_3$ may occur.
LSI is close to zero: Borderline scale potential.

Corrosion rate calculations

The rate of corrosion of Brass in water is influenced by several factors related to the water, including pH, temperature, electrical conductivity, dissolved elements, and water movement. Additionally, the composition of the alloy, the processing method (such as extrusion or rolling), heat treatment, and surface state play a role.[23]. By using the following relation corrosion rate can be calculated [24]:

$$CR = \frac{Wx365}{\rho xAxT} \quad \text{(cm/year)} \quad (3)$$

Where:
W = weight loss (g)
365 = number of days per year (day/year)
A = the area of the specimen exposed to the solution (cm^2)
ρ = the metal density ($8.73 g/cm^3$)
T = exposure time of the specimens (day)

Results and Discussion

The results shown in Table 3 are the results when the experiments were run under close system Visual observations revealed that the color of the Gabraun water remained stable, attributed to the inhibitor's effect, which slows the reaction between copper (Cu) and sulfate (SO4). However, the specimens' color darkened upon immersion in both Gabraun water with and without the inhibitor, as illustrated in Figures 3 and 4. Moreover, it is assumed that the decrease of the amount of Oxygen in this system was also played role in these results mentioned above.

Table 3: Results of the corrosion tests for brass specimens in close system

No Specimen	Weight before immersion (g)	Weight after immersion (g)	Weight loss (W) (g)	Area (Cm^2)	Corrosion rate cm/year	Weight of Inhibitor (g)
1	13.2626	12.8555	0.4071	7.634	0.0455	0.2500
2	13.3606	12.9566	0.4037	7.854	0.0438	0.5041
3	13.6774	13.2637	0.4137	7.854	0.0449	0.7508
4	13.4591	13.0520	0.4071	7.854	0.0442	1.0003
5	13.3042	12.9036	0.4006	7.656	0.0446	1.2500
6	13.3049	12.9043	0.4006	7.854	0.0435	1.7510
7	12.9116	12.5225	0.3891	7.540	0.0440	2.0063
8	11.6276	11.2774	0.3452	7.068	0.0416	Without
9	12.6432	12.2618	0.3814	7.539	0.0432	Without
10	12.6423	12.2563	0.3860	7.539	0.0436	Without
11	12.6503	12.2684	0.3819	7.586	0.0429	Without

Fig. 3: Specimen after the immersion in Gabraun water with inhibitor

Fig. 4: Specimen after the immersion in Gabraun water without inhibitor

Fig. 5: Weight loss (g) vs. weight of Inhibitor (g)

According to the figure 5, when the maximum Inhibitor weight (2.0063g) used, the lowest weight loss (0.3891g) produced, while the highest weight loss is 0.4137g when the weight of Inhibitor is 0.7508g.
Referring to figure 6, it is obvious that the lowest corrosion rate is 0.0435 cm/year when the weight is 1.751g, while the highest corrosion rate is 0.0455 cm/year when the weight of the Inhibitor is 0.25g.

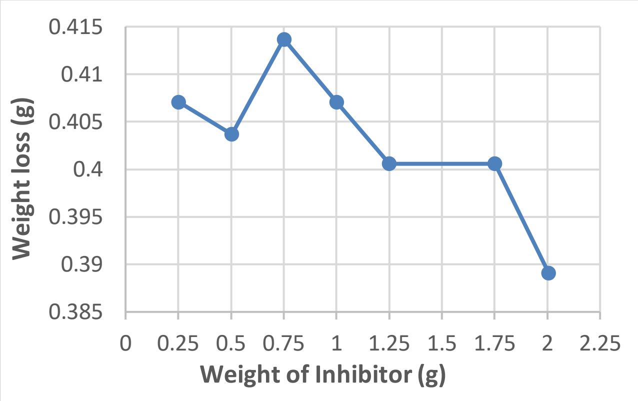

Fig. 6: Corrosion rate vs. weight of Inhibitor (g)

Referring to Table 3, it is clear that specimen's number 2, 3, 4 and 6 have the same exposed areas to the environment which is 7.854cm^2 and it is obtained that the lowest corrosion rate for the specimen number 6 which is 0.0435cm/year and the inhibitor amount is 1.751g, while all the results of these specimens are very close to each other. This shows that this inhibitor has not big effect because there is no big differences in corrosion rate between all the specimens tested with inhibitor and without inhibitor. So that this inhibitor is not useful for decreasing the corrosion rate of the Brass in this environment but it is useful for stopping or retarding the reaction between copper and Sulfate. The specimen color shown in figure 4 immersed without inhibitor is a little bit darker than that immersed with inhibitor shown in figure 3, this is because the inhibitor has a low effect on the specimen's corrosion rates.

Table 4: chemical composition of Brass (wt.%) after immersion in Gabraun water with inhibitor

Cu wt.%	Zn wt.%	Si wt.%	Pb wt.%	Al wt.%	Fe wt.%	Ni wt.%	Sn wt.%	Co wt.%	Mo wt.%	Au wt.%	Pt wt.%	Cd wt.%	Ca wt.%
57.241	36.570	1.1031	2.5242	1.5503	0.3737	.02763	0.2653	0.0082	0.0112	0.0207	0.0294	0.0029	0.0232

The findings shown in Ttable 4, clarifying that the Zinc (Zn) amount after the immersion in Gabraun water with inhibitor is decreased to 36.8 mass %, but the amount before the immersion the amount is 37.1 mass% as shown in table 1, this means that dezincification was occurred. Prior to immersion, the copper content (Cu) was 57.8 mass%, which decreased to 57.6 mass% after immersion. These findings suggest that zinc has leached out, resulting in the formation of a porous copper-rich structure with reduced mechanical strength, thereby indicating the occurrence of dezincification [23,25]. Dezincification is likely to occur in copper-zinc alloys containing more than 15% zinc [23,25]. It is known from the galvanic series ranking that Zinc is one of the highest reactive metals. This suggests that Zinc possesses a relatively weak atomic bond in comparison to other.

Table 5: chemical composition of Brass (wt.%) after immersion in Gabraun water without inhibitor

Cu wt.%	Zn wt.%	Si wt.%	Pb wt.%	Al wt.%	Fe wt.%	Ni wt.%	Sn wt.%	Co wt.%	Mo wt.%	Au wt.%	Pt wt.%	Cd wt.%	Ca wt.%
57.5	36.2	0.891	2.08	0.632	0.312	.0262	0.252	0.0114	0.0208	0.0272	0.0230	0.0029	0.0208

The obtained results in table 5, shown that the Zinc (Zn) amount after the immersion without inhibitor is decreased to 36.2 mass % while the amount before the immersion shown in table 1 is 37.1 mass%, this means that dezincification was occurred. Before the immersion the copper amount was 57.8 mass%, while after the immersion without inhibitor was found decreased to 57.6 mass%. These results indicate that Zinc leaches out, this means also dezincification is occurred.

From the above results, it is seen that the Zinc and copper amounts in table 5 is less than in table 4 and also less than in table 1, but there is not big difference between them. This means that the expired Esomeprazole inhibitor has a low effect on the Brass corrosion.

Table 6: Results of the brass specimens corrosion tests in an open system

No Specimen	Weight before immersion (g)	Weight after immersion (g)	Weight loss (W) (g)	Area (Cm^2)	Corrosion rate cm/year	Weight of Inhibitor
1	6.0373	5.8503	0.187	4.632	0.0344	0.2512
2	5.5958	5.4246	0.1712	4.241	0.0344	0.5066
3	5.5900	5.4195	0.1705	4.209	0.0345	0.7514
4	5.5013	5.3346	0.1667	4.398	0.0323	1.0043

The Gabraun water used in the brass corrosion tests turned dark blue, attributed to the reaction between copper (Cu) and sulfate (SO4), resulting in the formation of copper sulfate (CuSO4). These observations were made during an open system experiment conducted without inhibitors [19]. In this work, the findings in Table 6 were obtained from experiments run in open system with inhibitors. From the visual observations, the color of Gabraun water did not change during the period of the experiments, this is referring to the effect of the inhibitor (Expired Esomeprazole) on the reaction between copper (Cu) and sulfate (SO_4). The inhibitor (Esomeprazole) retarded the reaction between copper and sulfate, this is why no change in water color.

In addition, the results in table 6 can show the small differences between the weight loss and the corrosion rate for all the specimens, this means that the Esomeprazole inhibitor is not that inhibitor can give good results on the brass corrosion.

Although the actual results from the Brass experiments indicated that the Gabraun water sample was a corrosive medium, the Langelier Saturation Index (LSI) of +2.482 and the Pourbaix diagram for the copper–water system at 25°C in figure (1) show that the Gabraun water sample is not a corrosive medium.

Conclusion

1) There is no change in Gabruan water, this due to the effect of the inhibitor that retards the reaction between copper (Cu) and Sulfate (SO_4).
2) It is concluded that the decrease of the Oxygen amount in this close system experiments were also might be played role in these results.
3) The corrosion rates between all the specimens tested with and without inhibitor have not those big differences.
4) The Esomeprazol inhibitor has not good effect on corrosion rate.
5) Gabraun water is a corrosive media when used as electrolyte for testing Brass, this is opposite to the results of (LSI).

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