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A Thermodynamic Study of Inhibition of Copper Alloy Corrosion in Nitric Acid Using Laurel Leaf Extract

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ABSTRACT

The corrosion inhibition efficiency of copper alloy in HNO_3 solution at different concentrations (0.5, 1, 1.5, and 2 mol/L) was studied by adding bay leaf extract at different concentrations (1000, 2000, 3000, and 4000 ppm). The effect of concentration on the corrosion rate was studied in both the absence and presence of the inhibitor, in addition to choosing the acid concentration (1 mol/L) and varying inhibitor concentrations. The research also included studying the effect of temperature on the corrosion rate in both the absence and presence of the inhibitor. The weight loss method was used to calculate the corrosion rate. A negative heat of adsorption indicates that the reaction is exothermic. The adsorption free energy of the bay leaf extract ranged from 4.6 to 6.1 kJ/mol, and the results indicated that Langmuir adsorption was best. The results of this study showed that the inhibitor reduces corrosion in acidic media and that the inhibition efficiency increases with increasing inhibitor concentration and decreases with increasing temperature. The inhibition rate of the extract reached (92.31%) at an acid concentration of 0.5 mol/L, an inhibitor concentration of 4000 ppm, and a temperature of 298 K. This study showed that the inhibitor has high corrosion resistance. The results also showed that the bay extract exhibits inhibitory activity, likely due to its high viscosity and high levels of phenolic compounds and flavonoids, as indicated by the HPLC test.

Keywords: Corrosion, Copper Alloy, Nitric Acid, Weight Loss, Laurel Extract.

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دراسة ديناميكية حرارية لتثبيط تآكل سبيكة النحاس في حامض النتريك باستخدام مستخلص اوراق الغار

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الملخص

تمت دراسة كفاءة تثبيط تآكل سبيكة النحاس في محلول **HNO3** عند تركيزات مختلفة (0.5، 1، 1.5، 2 مول/لتر) وذلك بإضافة مستخلص الغار بتركيزات مختلفة (1000، 2000، 3000، و4000 جزء في المليون). و دراسة تأثير التركيز على معدل التآكل في كل من غياب ووجود المثبط، بالإضافة إلى اختيار تركيز الحمض (1 مول/لتر) وتركيزات المثبط المتغيرة. يتضمن البحث دراسة تأثير درجة الحرارة على معدل التآكل في كل من غياب ووجود المثبط. تم استخدام طريقة فقدان الوزن لحساب معدل التآكل. وتشير القيمة السالبة لحرارة الامتزاز إلى أن التفاعل هو عملية طاردة للحرارة. تراوحت قيمة امتزاز الطاقة الحرة لمستخلص الغار بين (4.6 كيلوجول/مول) و(6.1 كيلوجول/مول)، وأشارت النتائج إلى أن امتزاز لانجموير هو الأفضل. أظهرت نتائج هذه الدراسة أن المثبط يقلل من التآكل في الأوساط الحمضية وأن كفاءة التثبيط تزداد بزيادة تركيز المثبط وتنخفض بزيادة درجة الحرارة. بلغت نسبة التثبيط للمستخلص (92.31%) عند تركيز الحمض (0.5 مول/لتر) وتركيز المثبط (4000 جزء في المليون) ودرجة الحرارة (298 كلفن). من خلال هذه الدراسة، تبين أن المثبط لديه قدرة عالية على مقاومة التآكل. كما أظهرت النتائج أن مستخلص الغار لديه قدرة تثبيطية بسبب لزوجة المستخلص ومحتواه العالي من المركبات الفينولية والفلافونويدات، وذلك وفقًا لنتائج اختبار (**H PLC**).

INTRODUCTION

Corrosion is a general term for a wide range of processes that cause gradual, spontaneous damage and decay in metal materials during use.[1]. Brass, an alloy of copper and zinc, is known for its technical properties and has a wide range of uses, from mechanical engineering to medical applications and the arts. Certain factors cause the alloy to corrode in both copper areas, often beginning with the selective decomposition of zinc. [2-6]. The use of inhibitors is one of the scientifically proven and most economically effective ways to protect copper from corrosion.[7]. Organic compounds with higher molecular weight and heteroatoms (with lone pairs of electrons, such as N, S, and O) are commonly used as inhibitors. Unfortunately, many organic and inorganic inhibitors are toxic and non-biodegradable. As understanding of health and the environment grows, more emphasis is now being placed on the use of natural plant products. Plant extracts are organic substances containing proteins, polyacrylic acids, polysaccharides, alkaloids, pigments, etc. These compounds act as potential inhibitors for many metals in acidic environments. [8-13]. Laurel leaf extract contains many active compounds (alkaloids, phenols, amino acids, flavonoids, terpenes, sterols, carbohydrates and saponins). Alkaloids can be used as antioxidants and corrosion inhibitors. [14]. The present work aims to demonstrate the corrosion-inhibition properties of the aqueous extract of laurel using weight-loss measurements of a brass alloy in 1 mol/L nitric acid, and then to calculate several thermodynamic parameters and analyze the adsorption of the inhibitor on the brass alloy surface.

MATERIALS AND METHODS

Result of Sample

The components of copper alloys are determined by XRF (X-ray fluorescence as shown in Table 1.

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Table1: the chemical composition of the brass alloy.

Elements	Ni	LEC (Light Element Content)*	Zn	Cu
Wt %	0.01	4.15	36.40	Base
* LEC (Light Element Content) The nominal for the light element (Mg, Al, Si, P, S) content				

Sample Preparation

It is essential to create a homogeneous surface, as the alloy's surface condition plays a major role in its corrosion resistance. The sample was mechanically cut into equal pieces using a water jet, with dimensions of 2.5 cm in length, 2 cm in width, and 0.8 mm in diameter. Sandpaper was used to clean and polish the sample, then it was treated with acetone and distilled water and left to dry in a drying oven.

Measure Mass Cost

Changed concentrations of nitric acid (0.5, 1, 1.5mol/L), and 2 mol/L were produced and utilized as a corrosion medium. After being submerged in the corrosion medium for 1 hour, the metal samples were removed from the mixture, rinsed with distilled water, and allowed to dry. The solubility of clove oil and its extract in the corrosive medium was tested at various concentrations (1000, 2000, 3000, and 4000 PPM). The alloy's weight before and after immersion was used to calculate the weight loss (ΔW) in grams using the formula.

$W=W_i - W_f$ ….. (1)Δ

where ΔW (gram) represents the weight change, Wf and Wi represent the weight of the alloy after and before immersion.

With the weight loss approach, the corrosion rate was calculated based on equation (2) [15]. With more realistically simulated experimental settings than electrochemical approaches, the weightiness difference method—which is non-electrochemical—provides more reliable results for estimating corrosion rate and inhibition efficiency[16].

$$CR\left(\frac{mm}{y}\right) = \frac{\Delta W * 87.6}{AtD} \quad …. (2)$$

where ΔW (milligram) represents the weight change, A (cm2) surface area, t (hour) immersion time, and D (g/cm3) metal density.IE delay efficiency is calculated by equation (3) [17].

$$IE = \frac{CR - CRi}{CRu} * 100 \quad ….. (3)$$

CRi, CRu (mm/y): corrosion rates in the presence and absence of an inhibitor. The surface coverage is found from equation (4).

$$\theta = \frac{IE}{100} \quad ……(4)$$

Optical Microscopy

The metal specimens were studied with an optical microscope at 10X magnification of a Brass alloy immersed in (1mol/L) of nitric acid for an hour in the absence and presence of the inhibitor, as shown in Figure 1. Model (A) shows an optical micrograph of a polished Brass alloy with a smooth surface. Sample (B) shows an image of the metal after immersion in the corrosion solution. As for model (C), the optical microscopic image of the metal surface after immersion in a corrosion solution containing Laurel Extract at 4000 ppm is shown.

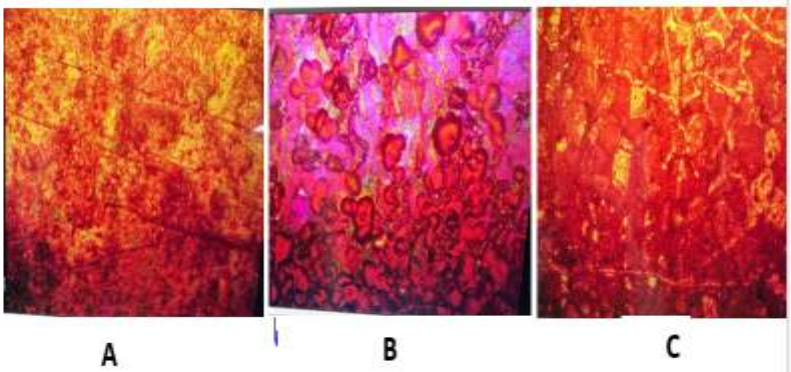

Fig. 1: photoscope images of low brass in the acid medium with magnification power (10x).

High Performance Liquid Chromatography (HPLC)

It was found that the laboratory-prepared Laurel leaf extract contains numerous active compounds. Table 2 shows the flavonoid and phenolic compounds present in the Laurel leaf extract according to the results of high-performance liquid chromatography.

Table 2: The flavonoids and phenolic composites present in laurel Leaf extract.

No	Reten t.(minu.)	Title	Concent. (ppm)
1	5.41	Qurcetine	9.8
2	6.77	Rutin	13.6
3	8.11	Kaempferol	10.8
4	9.01	Apigenin	7.8
5	11.25	Hydrobenzoic acid	8.8

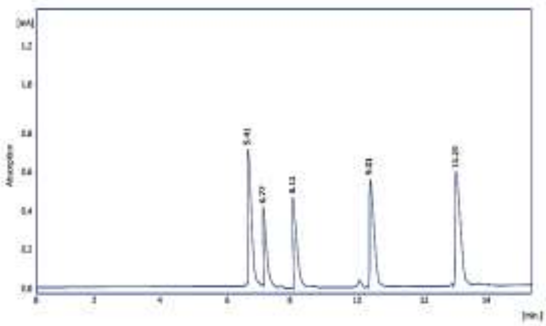

Fig. 2: HPLC chromatography chart of laurel Leaf Extra.

RESULTS AND DISCUSSION

Erosion Rate in the Presence and Absence of Inhibitor

In acidic solutions, corrosion rates can be determined using loss measurements at different concentrations (0.5, 1, 1.5, 2 mol/L) of Nitric Acid after an exposure period of one hour at a constant temperature (298K). It was found that the corrosion rate increased from 0.5 mol/L to 2 mol/L as the acid concentration increased. Figure 3 illustrates how the concentration of acid affects the pace at which copper alloy corrodes at a certain temperature. Additionally, at various temperatures (298, 308, 318, and 328K), the corrosion rate was computed. Research indicates that, as temperature increases from 298 to 328K, the corrosion rate of a copper alloy immersed in 1 mol/L nitric acid increases. Figure 4 shows the relationship between temperature and corrosion rate, where the corrosion rate increases with temperature.

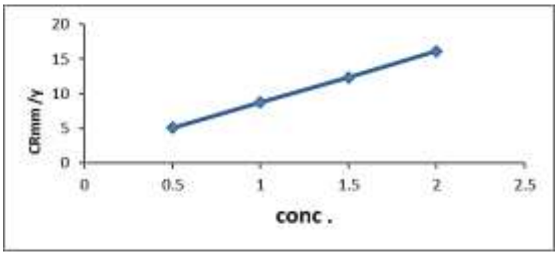

Fig. 3: The relationship between the corrosion rate of copper alloy and concentration of Nitric Acid at 298 K and time 1 hour.

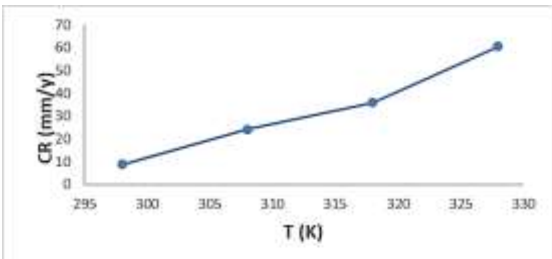

Fig. 4: The relationship between the corrosion rate of copper alloy in 1mol/L of Nitric Acid and temperature.

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The Effect of Inhibitor Effectiveness on Erosion Rates

The effect of the inhibitor on erosion rates generally indicates that the addition of the damper significantly lowers corrosion rates when submerged in solutions with varying acid concentrations (0.5, 1, 1.5, and 2 mol/L). Figure 5 illustrates that the corrosion rate decreases as the inhibitor concentration increases across various acid concentrations at a constant temperature (298 K), with the damper added at concentrations of 1000, 2000, 3000, and 4000 PPM. It significantly reduces the corrosion rate with increasing temperature, a phenomenon attributed to reduced interactions between the inhibitor molecules and the metal surface. [18] , as shown in Figure 6. It is believed that phytochemical components are responsible for oils' ability to prevent rusting [19].

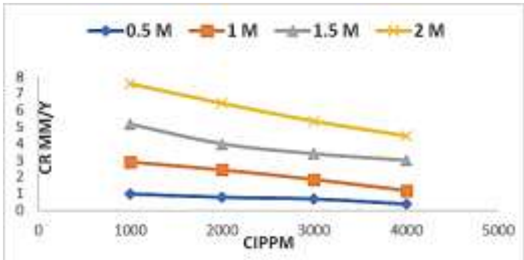

Fig. 5: The impact of varying the laurel extract content on the rate of corrosion of metal submerged in varying nitric acid concentrations at 298 Kelvin.

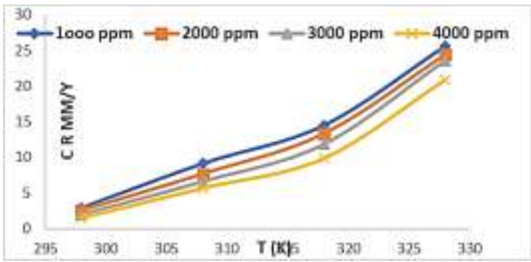

Fig. 6: Relationship between the corrosion rate of copper alloy and different temperatures in Kelvin at 1 mol/L Nitric Acid, varying concentrations of laurel extract and one hour time.

Effect of Inhibitor Concentration on Inhibition Efficiency

At a constant temperature (298 Kelvin), Figure (7) demonstrates that the inhibition efficiency rises with increasing inhibitor concentrations (1000, 2000, 3000, and 4000 ppm) at each acid concentration (0.5, 1, 1.5.2 mol/L). The highest inhibition efficiency (92.31) was achieved for laurel extract at a concentration of 4000 ppm and an acid concentration of 0.5 mol/L. The increase in inhibitor molecules adsorbed on the metal surface, which shields the metal from corrosion and prevents attacks on active sites, explains the rise in inhibition efficiency. [20].

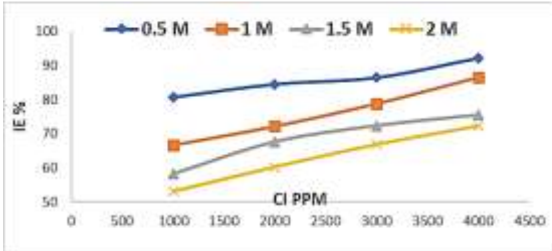

Fig. 7: The relationship between the inhibition efficiency and the concentrations of laurel leaf extract at varying Nitric Acid concentrations and temperature (298K) and time of one hour.

Effect of Temperature on Efficiency of Inhibition

The relationship between temperature and inhibition efficiency is depicted in Figure 8 for various inhibitor concentrations at an acid concentration of 1mol/L. For a solution containing a particular concentration of inhibitors, plot the inhibition efficiency of the inhibitor reduction with increasing temperature at that inhibitor concentration and at a specific acid concentration (1 mol/L). This means that the inhibition efficacy decreases at the highest temperature (328K) and the lowest inhibitor concentration (1000 ppm), corresponding to 57.5%.

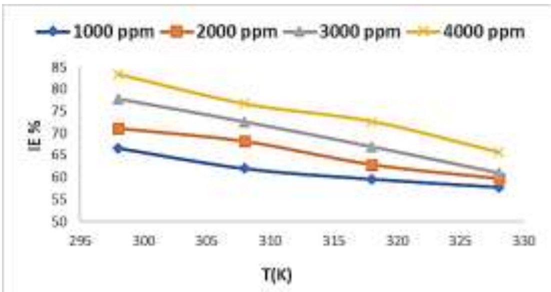

Fig. 8: The impact of temperature on the effectiveness of erosion inhibition of brass alloy submerged in 1mol/L Nitric Acid with varying amounts of laurel extract.

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Inhibitor Performance and Adsorption Interpretation

The adsorption of organic molecules on the metal surface determines how effective they are at preventing corrosion. It is evident that adding inhibitors lowers both the corrosion rate and the surface area that the inhibitor molecules θ cover. It is possible to investigate the adsorption mechanism of the Langmuir equation using the following equation by examining the connection between the inhibitor concentration (Ci) and the surface coverage θ determined from equation (4) [21].

Ci/θ =(1)/(Kads)+Ci …..(5)

The intercept line equals 1/k obtained by plotting the values of Ci/(θ) versus Ci in Figure 9. Table (3) shows the values of KL, the enantiomer constant at 1mol/L of Nitric Acid at different temperatures (298, 308, 318, 328 K). According to the data, the inhibition efficiency increases with increasing inhibitor concentration. The inhibitor is adsorbed onto the metal surface, blocking the acid-attack sites and preventing corrosion. [12]. We observe that the equilibrium constant Kads decreases with increasing temperature when the inhibitor concentration is held constant. According to the Freundlich equation (6), the computed corrosion rate values can be used to investigate the adsorption process. [22].

Ln θ =ln kf +n ln Ci …..(6)

Figure (10) shows a plot of Ln θ values versus ln Ci, indicating that the slope is equal to n values of Freundlich constants, and the intercept is equal to ln Kf. [17]. The Frendluch isotherm does not fit this system well, whereas the Langmuir isotherm fits the adsorption system better, as evidenced by its higher correlation coefficient. Based on the low values of the correlation coefficient R2, as represented in Table 4.

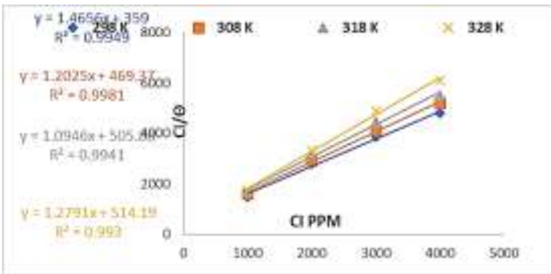

Fig. 9: The relationship between Ci/θ and Ci concentrations of laurel extract at varying temperatures of adsorption of the extract on the surface of brass alloy immersed in 1mol/L of Nitric Acid by the Langmuir method.

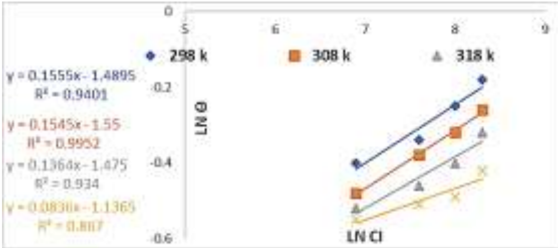

Fig. 10: Freundlich adsorption isotherm of laurel extract on metal immersed in (1 mol/L) nitric acid.

Table 3: values of the correlation coefficient R2 and Langmuir adsorption constant KL (ppm) for the Langmuir adsorption equation of laurel extract in 1 mol/L of Nitric Acid at various temperatures.

T (K)	KL(ppm)	R2
298	2.78×10-3	0.9949
308	10-3×2.13	0.9981
318	1.98×10-3	0.9941
328	2.78×10-3	0.9930

Table 4: Values of the Freundlich adsorption constant kf (ppm), slope n" and correlation coefficient R2 for laurel extract in 1mol/L Nitric Acid and different temperatures.

T (K)	K_f(PPM)	n"	R^2
298	0.225	0. 1555	0.9401
308	0.212	0.1545	0.9952
318	0.228	0.1364	0.934
328	0.321	0.0836	0.867

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Thermodynamic Adsorption

Understanding the type of adsorption, determining the nature of the system, and describing the molecular organization all depend on thermodynamic investigation.

From the values of the balance constant for (Kads) presented in Table (3), the free energy of (ΔGo) can be calculated from equation (7) [(23)].

$\Delta G^{O} = -RTln\ (k_{ads} \times 55.5)$ ....(7)

T= temperature in Kelvin, R= gas constant value. 55.5 represents the molar concentration of water in the solution.

When using the Van't Hoff equation and diagram, the relationship between (lnKads) versus (1/T), the slope value in Figure (11) represents (-ΔHo/ R), and from it we find ΔHo. , and the entropy value (ΔSo) was calculated from the Gypsum equation (8) [(24)].

$\Delta G^{o}=\Delta H-T\Delta S^{o}$ ....(8)

The results of the studied functions (ΔG°, ΔH, ΔS) are shown in Table 5.

Table 5: Thermodynamic functions of adsorption of laurel extract at different concentrations on the surface of copper alloy at different temperatures when immersed in 1 mol/L of nitric acid.

T(K)	G^{O}Δ (KJ/mol)	HΔ (KJ/mol)	S^{O}Δ (KJ/mol.K)
298	4.6	10.81-	0.0517-
308	5.5		0.0530-
318	5.8		0.0522-
328	6.1		0.0516-

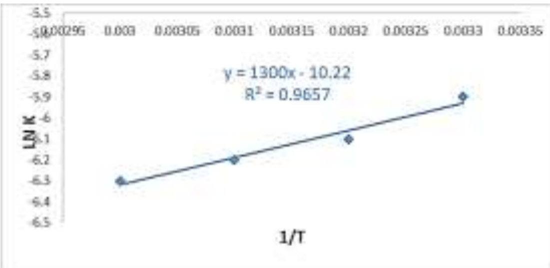

Fig. 11: The relationship between (LnK) and (1/T) for the adsorption of laurel extract in Nitric Acid (1mol/L) at different temperatures.

The values obtained (ΔH, ΔGO, ΔS) for the inhibitor at different temperatures listed in Table 5 can be discussed according to the following points:

- The negative value of enthalpy (ΔH) indicates that the adsorption process of the inhibitor is exothermic.
- The negative value of entropy (ΔS). It is shown that the system order increases during adsorption, as the molecules adsorbed become bound to the atoms of the adsorbed metal surface.
- The positive value of ΔGO indicates that the forces responsible for the adsorption method are spontaneous and of a physical nature.

CONCLUSION

1. The corrosion rate of copper alloy in Nitric Acid solution increases with increasing acid concentration.
2. With increasing temperature, the corrosion rate increases.
3. The highest inhibition efficiency was obtained for laurel extract (92.31%). Inhibition efficiency increases with increasing barrier concentration and decreases with increasing temperature.
4. Laurel extract effectively inhibits the corrosion of copper alloys at different concentrations of Nitric acid.
5. Through the thermodynamic values provided and free energy, it was found that the adsorption of molecules on the surface of the alloys is due to physical adsorption. Acceptance is an exothermic process that reduces randomness.
6. From the values (R2), it was found that the laurel extract is consistent with Langmuir.
7. Optical microscope images show the presence of alloys in (1mol/L) of Nitric Acid, as shown in the external appearance of metals and alloys after unloading. Among the calculated results, it was found that the inhibition efficiency of laurel oil was high, due to the proportions of phenols and flavonoids in the extract.

Note that the workplace is in the Chemistry Department Laboratory / College of Education for Pure Sciences / University of Anbar.

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Author contribution: The author contributed to the study.

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