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Investigation of the Inhibitive Effect of Tobacco Leaves Extract on Corrosion of Alpha Brass in Acidic Solutions

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Abstract :

This study was a trial to find an organic inhibitor that can to easily be prepared and applied , as well as non- toxic , cheep and available. In the present study an aqueous extract of tobacco leaves as corrosion inhibitor to protect α brass alloy in acid solutions (hydrochloric and nitric acid) at 1 M concentration, the inhibitor efficiency was evaluated using the weight loss method and microscopic test. Results obtained reveal that the extract of tobacco leaves plants at various concentration (0.5, 1.5, and 3) vol. % show significant corrosion inhibition during immersion of brass in acid solutions. The inhibition efficiency increased with increasing concentration of the inhibitor, but decrease with increase in exposure time. Inhibitor efficiency in 1M HCl and 1M HNO_3 at (3%) concentration of inhibitor reached to 75.64% and 73.17% respectively. Microscopic examination test shows the clearness of the metals surface which is under the corrosion medium and which contains inhibitor, from any kind of corrosion that can be found on those surfaces, which are under mediums completely empty from inhibitor, and also shows the clearness of that protect layer on the surface. The data of corrosion rate show that the way of doing this organic inhibitor is adsorbing and the inhibitor molecules are adsorbed on the metal surface according to Langmuir adsorption isotherm.

Main Words: Inhibitors, brass, tobacco leaves, corrosion, Langmuir adsorption isotherm.

الخلاصة

تمثل هذه الدراسة محاولة لإيجاد مثبط عضوي سهل التحضير والتطبيق , غير سام , رخيص الثمن ومتوفر بكثرة. تم استخدام مستخلص مائي من أوراق نبات التبغ في تثبيط تآكل سبيكة البراص α في محلول حامضي (محلول حامض الهيدروكلوريك ومحلول حامض النتريك) وبتركيز 1M. تم تقييم كفاءة المثبط باستخدام طريقة فقدان الوزن واختبار الفحص ألمجهري . أظهرت النتائج إن مستخلص أوراق نبات التبغ بتراكيز مختلفة (0.5, 1.5, 3 vol. %) يتمتع بقابلية جيدة على التثبيط لسبيكة البراص المغمورة في أوساط حامضية و تزداد كفاءة التثبيط مع زيادة تركيز المثبط وتقل مع زيادة فترة الغمر. وصلت كفاءة تثبيط تآكل سبيكة البراص α في محلول حامض الهيدروكلوريك و في محلول حامض النتريك إلى (75.64%) و (73.17%) على التوالي عند تركيز (3%) من المثبط. اظهر اختبار الفحص ألمجهري خلو سطح سبيكة البراص المتعرض للمحلول الأكال الحاوي على المثبط من مظاهر التآكل المتواجدة على سطح النماذج المغمورة في الوسط الأكال الخالي من المثبط ووضوح طبقة الحماية الممتزة على السطح. بيانات معدلات التآكل بينت إن آلية عمل المثبط العضوي هي الامتزاز(Adsorbing) وان جزيئات المثبط تمتز على سطح المعدن طبقا إلى علاقة لانكمير للأمتزاز Langmuir (Adsorption Isotherm).

الكلمات الرئيسية : المثبطات , سبيكة البراص ألفا, أوراق نبات التبغ , التآكل , علاقة لانكمير للأمتزاز.

1. Introduction

Copper and its alloys have a long history of use in industry applications and continue to be widely used for many purposes despite the emergence in recent years of other materials that have technical or economic attraction [1]. These related to their corrosion resistance, mechanical workability, excellent electrical and thermal conductivies and resistance to micro-fouling [2].Copper alloys have gained increasing importance for applications sea water environments such as heat exchangers of power plants. Scale and corrosion products cause a decrease in the heating efficiency of the equipment, which is why periodic descaling and cleaning in acid-pickling solutions are necessary. In order to reduce the corrosion of the copper alloys during such surface treatments, a corrosion inhibitor is typically added [2]. The corrosion resistance of copper alloys has been attributed to the

formation of protective films of cuprous oxide (Cu_2O) and to the doping of Cu_2O layer with ions such as iron. This doping reduces the ionic or electronic conductivity of the film, improving the corrosion resistance. Since corrosion is one of the main concerns in the durability of materials and structures, many studies have been carried out to develop an effective means of corrosion control to prolong service life of existing structures and minimize corrosion damage in new structures. Even though corrosion inhibitors are the most effective and flexible means of corrosion control. The selection of an appropriate inhibitor for a particular system is actually complicated due to specificity of inhibitors and great variety of corrosion – related applications [3][4]. In order to improve corrosion protection of copper and copper alloys, numerous advanced treatments, including develop environmentally friendly corrosion inhibitors to replace the traditional inorganic corrosion inhibitors, such as chromates and lead, which have significant health, safety, and environmental concerns. Both are listed as persistent, bioaccumulative, and toxic (PBT) chemicals [5]. Because PBT chemicals do not readily break down or decrease in potency in the environment, they accumulate and have greater potential to cause long-term human health or ecological problems. In view of this , there is a need to develop new ecofriendly corrosion inhibitors, extracts from tobacco leaves plants show excellent corrosion inhibition properties for several metals [6][7]. The tobacco plant is a virtual chemical factory with over 4,000 compounds. Tobacco is currently being evaluated as a production system for antibiotics, sugars, industrial enzymes, and anti-cancer [8, 9, and 10]. Some of the tobacco constituents show remarkable corrosion inhibitive properties. Tobacco extracts represent a major new initiative in the corrosion inhibition market with the following potential advantages:

- Low cost and high effectiveness.
- Environmentally acceptable.
- Low toxicity.
- Readily available and renewable.

Corrosion inhibitors have been studied for many years [11] [12]. Many types of organic compounds have been found to act as inhibitors [13] [14], but most of these compounds have remained as laboratory data. One reason seems to have been that the cost of manufacture of these compounds is generally too high for the corrosion market. In contrast, tobacco extracts can be prepared at very low cost, especially if simple extract procedures can be used and scrap plant parts can be used without curing. Effective adsorbing inhibitors include aliphatic and aromatic amines, sulphur-compounds such as thiourea and substituted thioureas, carboxylic acids and their salts, aldehydes and ketones as well as numerous other organic substances. These substances exist either in the charged state or as neutral entities that are readily polarizable. Thus, in addition to the high surface adsorbability of N-, S- and O-compounds, the metal surface charge may be expected to affect adsorption. Synergistic (and antagonistic) effects are often found with mixtures of inhibitors and these effects may be related to the charge in the electrical double layer (edl) present between an electrode (the metal surface) and its environment [15]. Tobacco products contain a high concentration of alkaloids, fatty acids and N-containing compounds, many of which might be anticipated to exhibit electrochemical activity. Some 4,000 individual constituents have been identified in tobacco but electrochemical activity, requiring the presence of polarizable nitrogen, oxygen and sulphur atoms, is likely only with a limited number of constituents [16]. Poly nuclear aromatic hydrocarbons also may be electrochemically active due to their fused benzene ring system with its attendant charge dislocation. These tobacco constituents with anticipated electrochemical activity include terpenes, alcohols, polyphenols, carboxylic acids, nitrogen-containing compounds, and alkaloids. Although there are a few reports on the inhibitive properties of compounds that bear some resemblance to the constituents of tobacco, namely organic amines, 3-amino-5-heptyl-1,2,3-triazole (AHT), benzoyl allyl alcohol, pyrazole derivates and macro cyclic compounds [17][18].

2.0 Experimental work

2.1 Samples preparation

The material used in this work is brass. Analysis of this material was carried out using (spectrometer DV. 4) in AL- Nasser company. Table (1) shows the chemical composition of α brass alloy. The cross sectional surface area of each sample metal depends, of course on the diameter of the stock rod. However, the brass samples were cut into circular shapes of 16 mm as diameter and 3 mm as thickness, and a total surface of 565 mm^2. The samples were examined carefully to check for rough edges, which could influence the corrosion monitoring process. Small whole of 2 mm diameter was drilled in each sample for holding. The surfaces of both the cut ends and the body were slightly polished to remove trace of contaminants and to achieve a flat surface at both the cut edges, then the cut samples were degreased in alcohol. This was carried out to improve the adhesion of the epoxy mounting resin to the metal so as to reduce the tendency of the metal from experiencing crevice corrosion at the edge of the mounting resin. Polishing of the samples was done using SiC papers. The samples were ground on successively smaller grades of SiC paper from 220 to 1200 grit using water as lubricant on rotating grinding wheels. After polishing, the samples were washed in deionized water and dried. Then they were kept dry (in a dessicator) until they were needed to be used.

2.2 Inhibitor preparation

The inhibitor used in this experiment was derived from natural tobacco dried leaves. Extract was prepared then added to the test solution for the experiments. Tobacco dried leaves were obtained from a market and , 4 g of this material was immersed in 15 ml distilled water, boiling off the water in order to extract the active ingredients. Later the resultant mixture was filtered. This left a dark yellow residue, which represents tobacco leaves extract.

Tobacco leaves extract weighed and added to the test solution. The concentrations of the tobacco leaves extract were expressed as vol. / vol. percentage.

2.3 Weight loss corrosion test method

In the weight loss experiment, four beakers (250 ml) containing 1M HCl solutions with 0, 0.5, 1.5, and 3 vol. % tobacco leaves extractor as additives respectively as a first group. Whereas the second group contains 1M HNO_3 solutions with the same additives indicated in group one. All beakers were placed in thermo stated water bath maintained at 30 ^oC. The brass samples were suspended in the beakers with the aid of special hooks. The samples were retrieved from their corroding solutions at one hour material for 10 hours due to the reactivity of brass in HCl, HNO_3 solutions. Each set of samples was dipped into saturated ammonium acetate solutions at room temperature, to terminate the corrosion reaction. They were washed by distilled water and dried in acetone and finally in an oven maintained at 80 ^oC. The weight loss of brass samples was evaluated in grams as the difference in the weight of the samples before and after the test [19]:

$$\mathbf{W = (W_i - W_f) \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 1}$$

Where W= weight loss of sample.

W_i = Initial weight of sample.

W_f = Final weight of sample.

Each reading reported is an average of three experimental reading recorded to the nearest 0.0001 g on a Sartorius electronic balance. The inhibition efficiency (I.E %) of tobacco extract was calculated using the formula [20].

$$\mathbf{I.E\% = \{1- W_1/ W_2\} *100 \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 2}$$

Where W_1, and W_2, are the weight losses (mg) for brass in the presence and absence of inhibitors respectively in HCl and in HNO_3 solutions at the same test conditions. The degree of surface coverage, Θ is given by equation (3), [21]:

$$\mathbf{\Theta = \{1- W_1/ W_2\} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 3}$$

The corrosion rate of brass alloy in different HCl and HNO_3 concentration solutions was determined for a ten hours immersion period from weight loss using the formula [22]:

Corrosion Rate (mpy) = 534 w/ *DAT* …….. 4

Where: mpy = mils per year
W = weight loss (mg)
D = density of sample g/m^3
A = area of sample (sq. in.)
T = exposure time (hrs.)

3. Results and discussion

3.1 Fourier transform infrared test (FTIR)

Fig. 1 illustrates the results of FTIR spectroscopy measurement of tobacco leaves extract as inhibitor material used in this research. This figure shows that inhibitor material contains many active groups that are rich in alcohols , alkane, alkenes, esters compounds and hydrocarbon groups. Most of these materials have inhibitor properties. Further, the existence of double bonds in this inhibitor tends to improve inhibitor action. This result is in a good agreement with those given by Saleh et al. [23].

3.2 Weight loss corrosion test method

3.2.1 Effect of tobacco extracts on the corrosion of brass in 1M HCl

Fig. 2 shows the corrosion of α-brass alloy in (1M HCl) with and without tobacco extract concentrations (0.5, 1.5, and 3) % v/v. The results obtained indicate that the weight loss of samples immersed in HCl solutions without inhibitor increased with increased time continually, because the existence of hydrogen ions which tend to electrons consumption and corrosion accelerated. This result is in consistence with others [15]. The same Figure show a significant decrease in weight loss with a various concentration of additives (tobacco extracts) because tobacco extracts contain many organic active groups which can adsorb on the metal surface and block the active sites on this surfaces and thereby reduce the corrosion rate in acid environments.

Fig. 3 shows that the extent of decreasing in weight loss was depend on concentration of additives; it means height inhibition efficiency at higher additives concentration of 3% v/v. It was expected that most of sample surface would be coverage by inhibitor molecules

3.2.2 Effect of tobacco extracts on the corrosion of brass in 1M HNO_3

Fig. 4 shows the corrosion of the α-brass in (1M HNO_3) with and without tobacco extract concentrations (0.5, 1.5, and 3) % v/v. It seems clear that the sample immersion in 1M HNO_3 without inhibitor suffer a great weight loss, because oxidizing acids such as an nitric acid and acids containing oxidizing agents causing a dramatic increase in the rate of metal loss of cupper alloys ,especially brass alloy [24], but when tobacco extract added to the same acid solution with various concentration , the weight loss decreased with tobacco extent depending on the concentration of the additives, and on this basis we suggest the mechanism of physisorption of the inhibitor on the metal surface . Fig. 5 illustrates the variation of the inhibition efficiency IE% verse the concentration of the additives. It seems clear that the inhibition efficiency increases with increasing concentration of the inhibitor; this is due to the adsorption of more inhibitor molecules on the metal surface, However inhibition efficiency increases with increasing of inhibitor concentration. This is well known that adsorption is a function of inhibitor concentration that as concentration rises; the adsorbed quantity increases [25].

3.3 Microscopic test

The light optical microscope was used to evaluate the change in the surface formation caused by contact with the test solution, and to monitor the effect of adding inhibitor to test solution.

Fig. 6a shows the surface of blank sample after immersed in 1M HCL solution without inhibitor, it seems clearly the dense pitting corrosion with different sizes due to surface corrosion. Whereas Fig. 6b shows the sample surface after immersed in 3 vol. % tobacco extract with 1M HCl. It is obvious the large differences in both cases due to the activity of adsorbed inhibitor layer at the sample surface which protected against corrosion. Figure 7a shows microscopic test of the surface of brass sample after immersion in 1M HNO_3 solution for 10 hrs. at room temperature. The topography reveals that, the surface is strongly damaged in the absence of the inhibitor (active corrosion). While, Figure 7b shows image of the surface of another brass sample after immersion for the same time interval in 1M HNO_3 solution containing 3 vol. % tobacco extract. The topography reveals that, there is a decrease in the corrosion sites and pits over the surface of the brass; it appears that the surface of the specimen is covered greatly due to formation of surface film of the inhibitor. From these observations we can say that this natural organic inhibitor give a good inhibition effect for the brass alloy.

3.4 Adsorption isotherm

It is of generally view that inhibition of metals in acidic solution results from the adsorption of molecules or ions of the inhibitor on the metal surface. The action of organic inhibitors depends on the type of interaction between the substance and the metallic surface. This could cause a change either in the electrochemical process mechanism or on the surface available to the process. The decrease in inhibition efficiency with decreasing inhibitor concentration, suggest weak adsorption interaction between brass and the inhibitor molecules which is physical in nature [26].

The Langmuir adsorption isotherm may be expressed as [22]:

$$\Theta = K C / (KC + 1) \quad \text{.....5}$$

Where K is the equilibrium constant for the adsorption process, C is the concentration of the inhibitor and Θ is the surface coverage, 1 when inhibitor efficiency is 100%. Rearrangement of the Equation 5 yields to:

$$C / \Theta = (1 / K). C \quad \text{.....6}$$

It was found that Figures 8 and 9 (plot of C/ Θ) versus C gives straight lines with slop, practically equal to unity, indicating that the adsorption of compound under consideration on brass / acid solution interface obeys Langmuir adsorption isotherm. The deviation of the slope from unity is attributed to the difference in the rate of interaction between the adsorbed species on the metal surface. The interaction between the adsorbed species is not taken into account during deviation of the Langmuir isotherm equation, while the interaction between adsorbed organic molecules, with polar atoms or groups on the anodic and cathodic sites of the metal surface plays a crucial role. This interaction may be either mutual repulsion or attraction [27][28].

Finally, from the result of weight loss against time Figures 2 and 4, corrosion rate, and percentage inhibitor efficiency (Table 2) for metal dissolution was inhibited to a comparative degree. The inhibition action of leaves, fruits, bark of trees and plant has been attributed to tannins and nitrogenous compounds in the extracts. Both groups have been found effective for corrosion inhibition and inhibitors of high molecular weight (carbon atom 12 and above) are better inhibitors than the methyl derivatives [29]. It may be reasonable to suggest that corrosion inhibition by both additives may be due to bulky nitrogenous organic compound or tannins, which contain polar groups. For tobacco extracts, these may have effected inhibition through nitrogen bond on metal surface with the formation of complex ions on the surface of the metal.

4. Conclusions

From the present work, the following conclusions can be drawn:

- Results show that inhibitor efficiency increased from 33.3% to 75.6% in case of additive concentration (Tobacco extract) 0.5 and 3% respectively to 1M HCl. While inhibitor efficiency increased from 48.7% to 73.1% in case of additive concentration 0.5 and 3% respectively to 1M HNO_3 at the same other test conditions.

- The rate of corrosion of the α brass in HCl and HNO_3 solutions is a function of the concentration of tobacco leaves extract.
- The inhibition efficiency increases with increasing additive concentration. Inhibition though generally was obvious at an optimum concentration 3%. This may be due to protonation of the hydrogen evolution process or the formation of un-soluble complex on α brass surface by molecules of tobacco leaves.
- Tobacco leaves extract is a corrosion inhibitor for α brass in HCl, HNO_3 solutions and can be used to replace toxic chemicals.
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5. References

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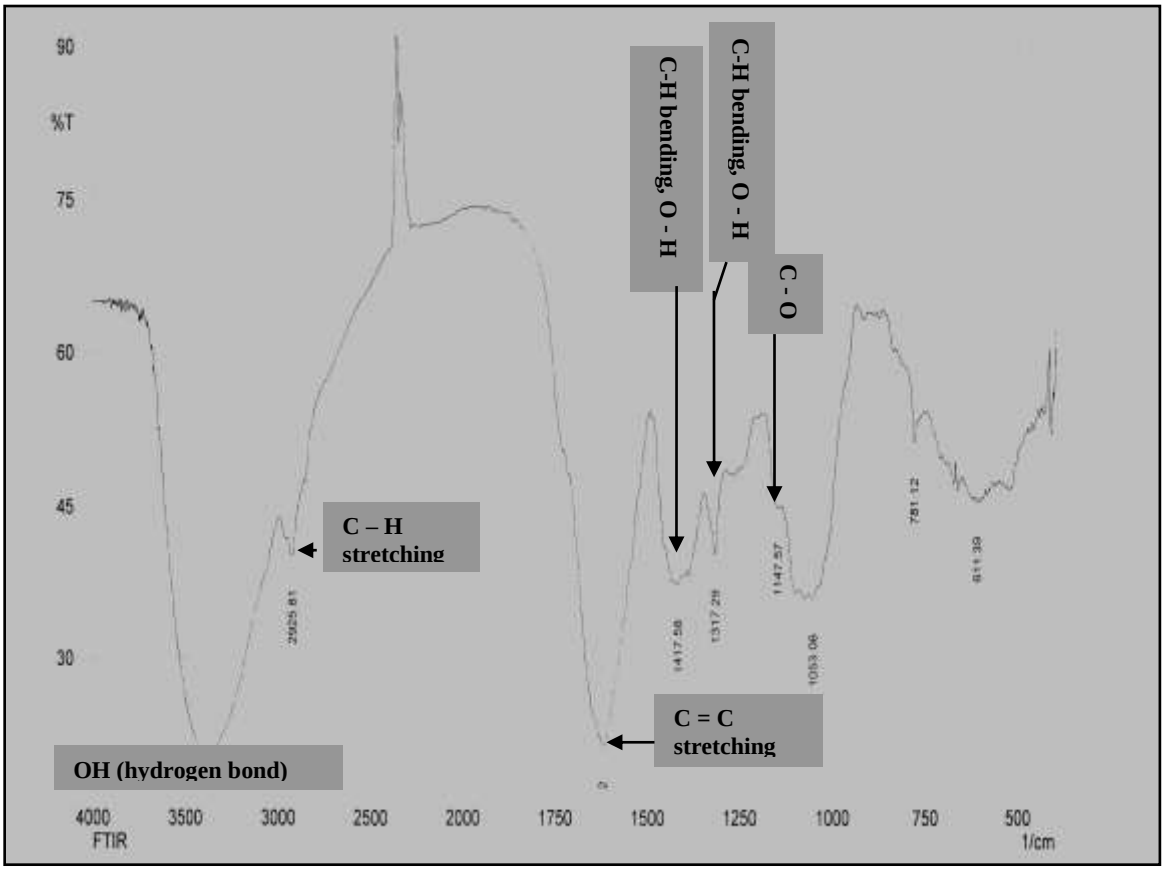


Transmitting %

Wave number (cm^{-1})

Fig. 1: FTIR pattern for tobacco leaves.

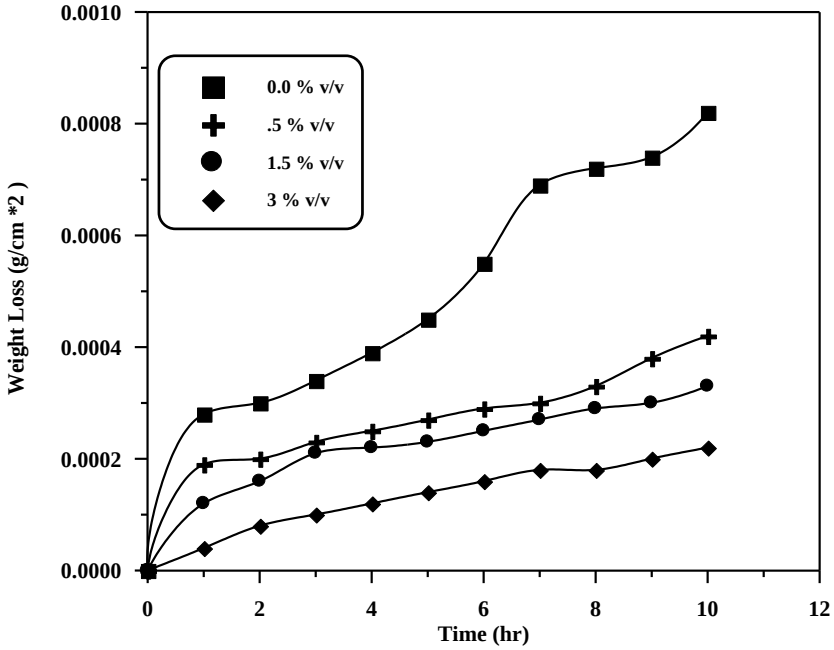


Fig. 2: Variation of weight loss with time for brass in 1M HCl solution containing different concentrations of tobacco extracts.

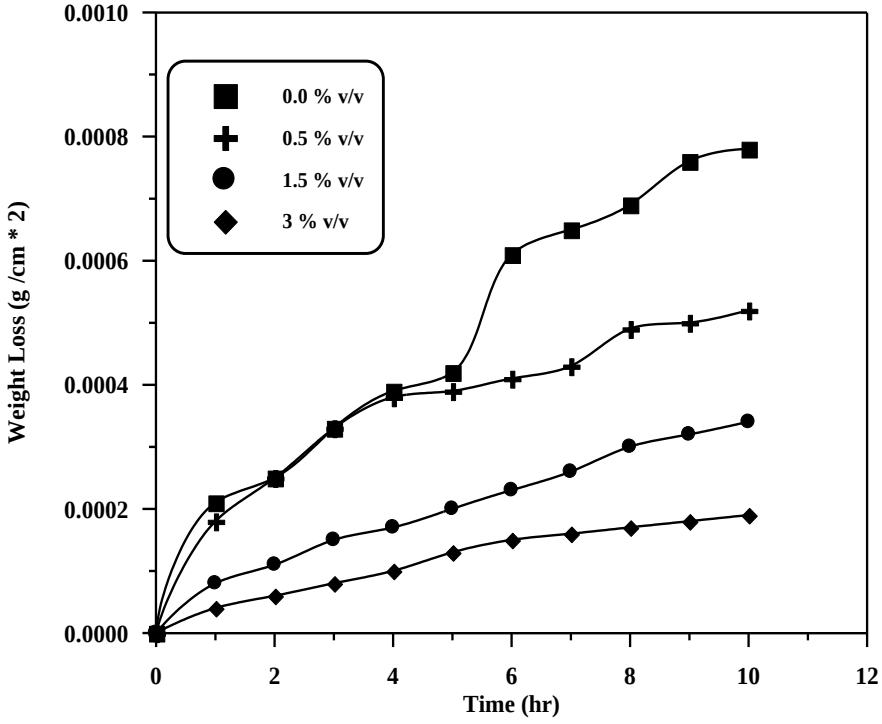


Fig. 3: Variation of inhibition efficiency with concentration in 1M HCl solution.

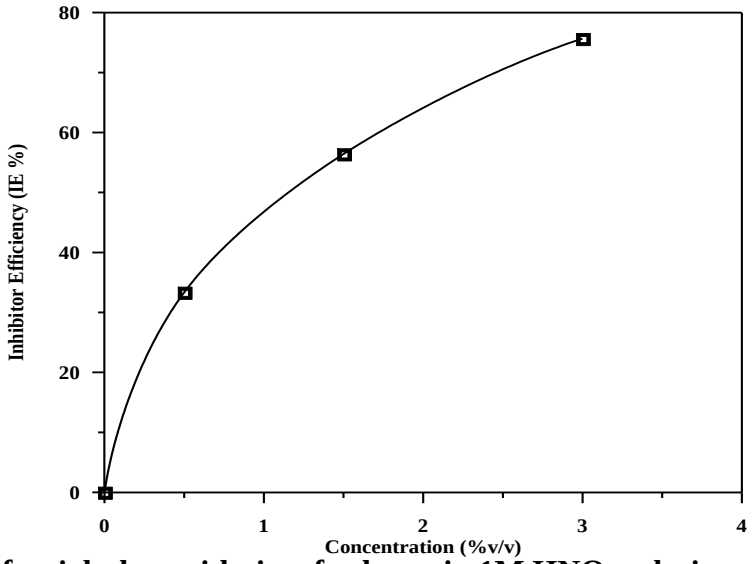


Fig. 4: Variation of weight loss with time for brass in 1M HNO_3 solution containing different concentrations of tobacco extracts.

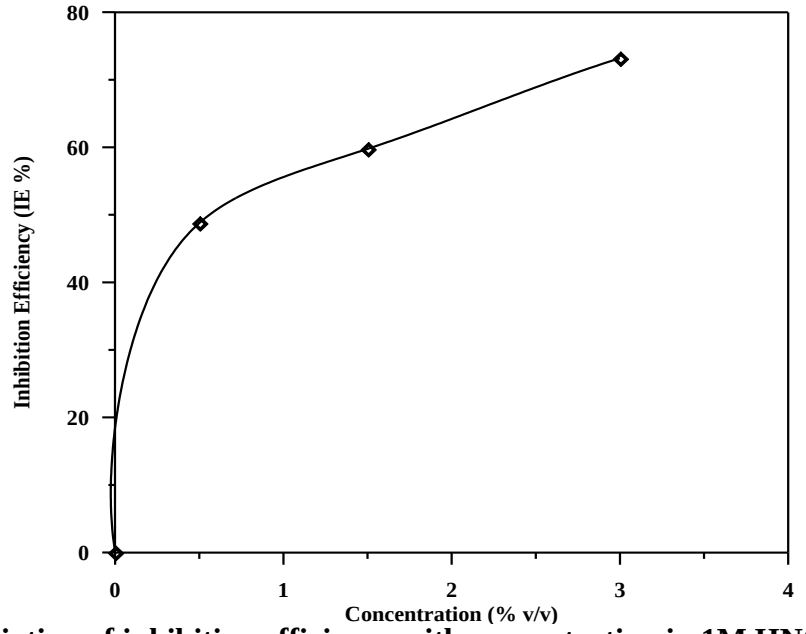
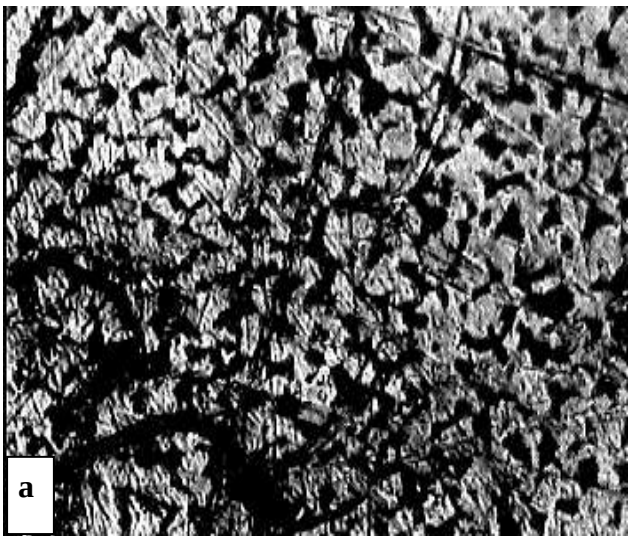


Fig 5: Variation of inhibition efficiency with concentration in 1M HNO_3 solution.



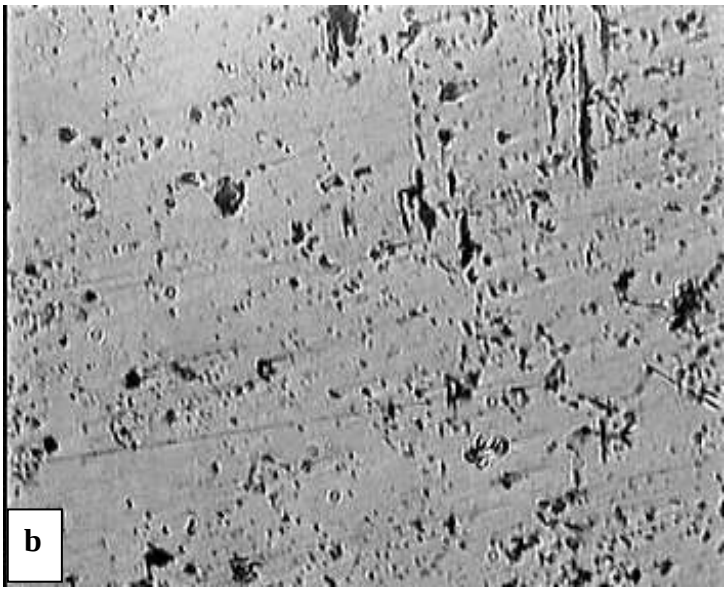


Fig.6: L.O.M. images of brass immersed in HCl solutions (a = 1M HCl, b = 1M HCL+3% tobacco extracts for ten hrs. (Magnification 400X).

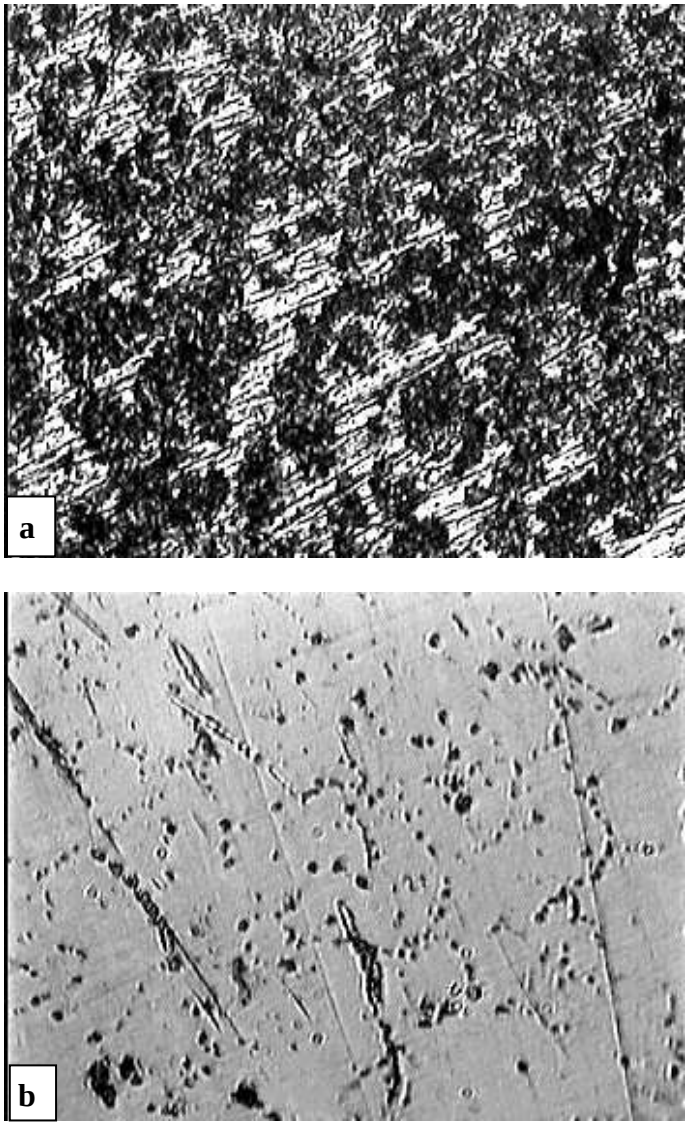


Fig.7: L.O.M. images of brass immersed in HNO_3 solutions (a = 1M HNO_3, b = 1M HNO_3+3% tobacco extracts for ten hrs. (Magnification 400X).

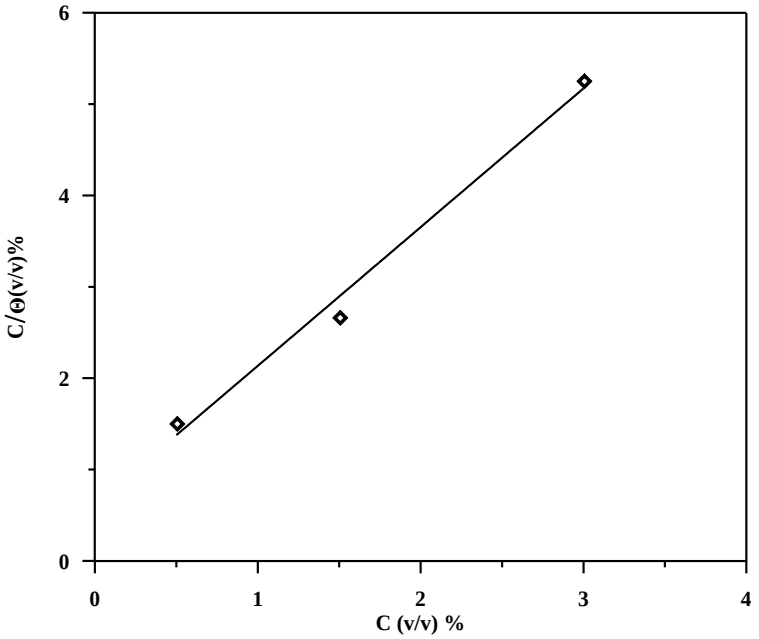


Fig.8: Langmuir isotherm plots for the adsorption of tobacco extracts on the surface of brass in1M HCL.

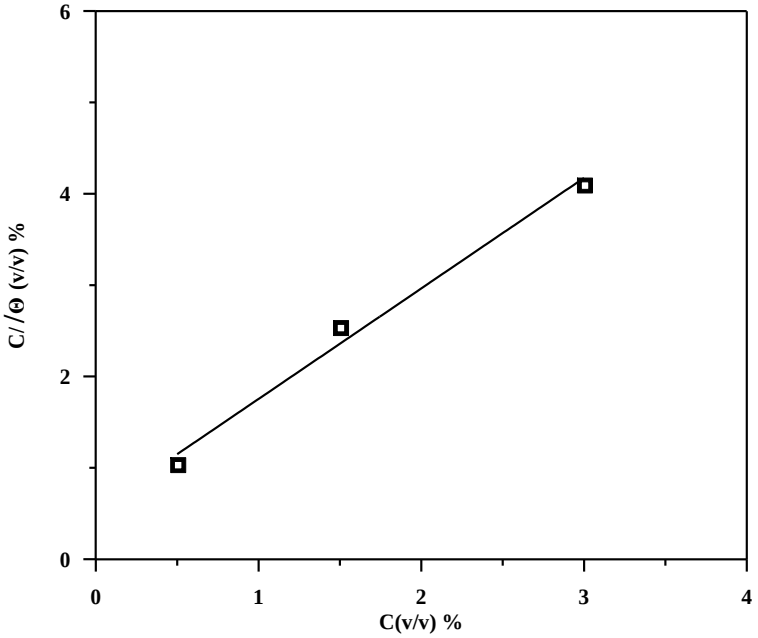


Fig.9: Langmuir isotherm plots for the adsorption of tobacco extracts on the surface of brass in 1M HNO_3

Table 1: Spectrochemical analysis of α- brass alloy wt. %.

Zn	Fe	Si	Mn	As	Ni	Sb	S	Cu
30.45	0.006	0.0036	.0088	0.005	0.0004	0.0084	0.007	Rem.

Table 2: Calculated values of % Inhibitor concentration, % Inhibitor efficiency, Corrosion rate (mpy) for corrosion of brass in (1M HCl, 1 M HNO_3) at 30 ^oC

Corrosion Medium	%Inhibitor Concentration	%Inhibitor Efficiency	Corrosion Rate (mpy)
1M HCl	0	-	29.994
	0.5	33.33	18.645
	1.5	56.41	12.970
	3.0	75.64	7.296
1M HNO_3	0	-	32.426
	0.5	48.78	14.592
	1.5	59.75	11.349
	3.0	73.17	8.917