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Utilization of Natural Nutmeg Oil in a 0.5 M H_2SO_4 Solution as an Environmentally Friendly Corrosion Inhibitor for Carbon Steel

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Abstract

In this research work we explore the potential of the Nutmeg oil as a natural, safe and effective corrosion inhibitor for X70 stainless steel in 0.5 M H_2SO_4 acidic corrosive medium. The corrosion rate of X70 carbon steel was assessed through a series of test, analysing its performance in the absence and the presence of the Nutmeg oil inhibitor under various conditions.

The study of the X70 steel, in the presence of Nutmeg oil inhibitor in different solution concentrations (0.125 $g.L^{-1}$, 0.25 $g.L^{-1}$, 0.5 $g.L^{-1}$, and 1 $g.L^{-1}$) in 0.5, M H_2SO_4 acidic medium, at 25 ^0C, after one hour of exposure confirmed that this natural substance 1 $g.L^{-1}$ showed the highest inhibition rate of stainless steel (1.585 $g.cm^{-2}.min^{-1}$). Such results demonstrated a direct relationship between the concentration of nutmeg oil, which is famous for preventing corrosion of stainless steel X70.

The temperature has a significant influence on the inhibition efficiency, with the highest efficiency 86 % at 30.8 °C and the lowest efficiency, is 14 %, at 70.0 °C. We found that there is a reverse relationship between temperature and corrosion inhibition rate.

Keywords: *Nutmeg oil, Corrosion inhibitor, Carbon steel, Inhibitor efficiency, Mass loss.*

1. Introduction

Metal corrosion is a natural and inevitable loss of metallic properties, of many metals, resulting from the oxidation process caused by atmospheric oxygen, particularly catalysed by humidity and ionic salts.

Corrosion inhibitors represent one of the most promising economic and technical approaches to reduce the corrosion process. They decrease the metal dissolution rate even at low concentrations of the medium. But certain inorganic substances, particularly chromates and their derivatives, have been reported to be highly toxic for use as corrosive inhibitor. Nevertheless, its name was altered because of its potential harm to humans. On the other hand, choosing natural products from various types of plants constitutes an important solution, especially in the industrial field. [1] As a result, the 21nd century has witnessed a positive shift in terms of green chemistry and its applications as corrosion inhibitors. The corrosion inhibitory potential of bioactives from different plant parts has been examined. Their extracts exhibit similar effect to that of synthetic inhibitors as described in various recent studies on green corrosion inhibitors [2,3].

Various phytochemical compounds were utilised, including both volatile and non-volatile ones, acids, and amino acids. [4] Among the natural substances exhibiting noteworthy corrosion inhibition effects in industrial applications, vegetable oils stand out and are discussed in the context of developing new “green corrosion inhibitors”.

In contemporary times, the utilisation of natural products as green corrosion stabilizers has emerged as dynamic and cutting-edge research field. Numerous vegetable extracts and vegetable oils are subjected to evaluate their efficacity in corrosion prevention. A range of plants, including trees and weeds, were investigated to identify diverse corrosion inhibitors. [5]

Recently, various research reports have highlighted the successful utilisation of natural vegetable extracts as corrosion inhibitors in diverse metals. These include Mitracarpus Hirtus [6], olive leaf [7], Ziziphora leaves [8], Aloysia citrodora [9], Euphorbia heterophylla L [10], Borage flower [11], Pineapple stem extract (Bromelain) [12], pigeon pea leaf [13], Cordia dichotoma seeds [14], Nutmeg oil [15], Swertia chirata [16], Citrus reticulata leaves [17], Brassica

oleracea L [18], Gloriosa superba seeds, Dracaena arborea Leaves, Gongronema latifoliuim, Jatropha Leaf Extract, Gush leaves [19].

In this study, we present the experimental results detailing the inhibition action of Nutmeg oil on X70 carbon steel in H_2SO_4 media, employing the straightforward weight loss method. [20]

2. Experimental

a- Materials

Sulfuric acid was purchased from Sigma-Aldrich. Infrared spectra were recorded on FTIR Tripoli University in the frequency range of 3900-450 cm^{1} using KBr. The precision balance is of model a Pye Unicam SP-3-300 infrared spectrophotometer.

The study of the corrosion phenomenon and its inhibition was conducted on pieces of carbon steel X70 . The chemical composition X79 steel is listed [21]: Fe (97.770), Mn (1.685), Si (0.245), Ti (0.190), Nb (0.067), Al (0.042), Cr (0.042), Ni (0.026), V (0.014), Cu (0.010), C (0.005), Mo (0.005), P (0.002), and S (0.001).

b- Inhibiting compound:

The natural substance inhibitor (Trimyristin) was obtained from a simple liquid solid extraction.

Trimyristin extraction: In a conical 20.00 g of ground nutmeg were gently refluxed in 50 mL of diethyl ether for 30 minutes. Then, the mixture was cooled at room temperature, decanted and cleaned with 20 mL diethyl ether. The superior solvent phases were combined, and solvent was evaporated. The nutmeg-scented yellow stable residue, (4.14 g), was recrystallized from ethanol to yield 1.02 g of white and odourless trimyristin solid, (Mp = 56-58 °C, lit 56.5 °C), (5 %).

The characterisation of the compound was simply realized from its TLC, Mp and by FTIR spectrum (provided in Figure 1), referring to the bibliographic data [22].

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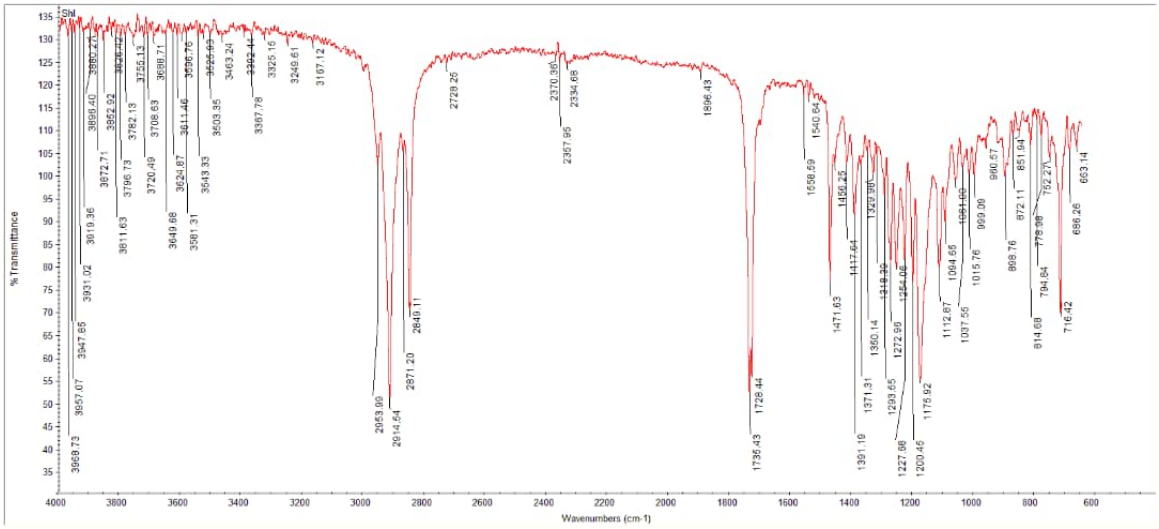

Figure 1. FTIR spectrum of Trimyristin. extract solid residue.

(2914-2871, C-H aliph sat), 1728, 1735 (C=O), 1227 and 1175 (C-O-C)

c- Polishing

This procedure includes cleaning the pieces of steel before using, by removing external contaminants by washing them with water and then acetone to ensure that there are no obvious defects or scratches and to make them smoother.

Preparation of the used solutions:

Corrosive medium: The corrosive medium used consisted of an aqueous solution of sulfuric acid at a concentration of 0.5 M (H_2SO_4), prepared from a commercial solution of sulfuric acid (H_2SO_4).

Corrosion study

In the absence of the inhibitor:

In a beaker containing 50 mL of the corrosion solution, 0.5 M (H_2SO_4), the steel samples were immersed vertically for 30 min, 60 min, 90 min, 120 min, 150 min and 180 min. The samples were kept at room temperature without stirring. At the end of the experiment, the samples were washed with distilled water, dried, and weighed to determine the amount of the loosed mass.

The dimensions of the pieces were measured to calculate the total surface area of each simple.

Preparation of inhibitor (mother solution):

b- In the presence of the inhibitor

The same previous method was used. The immersion time was kept for one hour, at different temperatures.

Solutions of different Nutmeg oil inhibitor concentrations (0.125 $g.L^{-1}$, 0.25 $g.L^{-1}$, 0.5 $g.L^{-1}$ and 1.0 $g.L^{-1}$) were prepared.

[12.5 g/L]: Add V = 12.5 mL of the inhibitor solution + H_2SO_4 and complete the volume with solution (0.5 M H_2SO_4 without inhibitor up to 100 mL.

[25 g/L] We add V = 25 mL of the inhibitor solution + H_2SO_4 and complete the volume with a solution (0.5 M H_2SO_4 without inhibitor up to 100 mL

[50 g/L] add 50 mL of inhibitor solution + H_2SO_4 and complete the volume with solution (0.5 M H_2SO_4 without inhibitor up to 100 mL

[100 g/L], We add 100 mL of solution (H_2SO_4 + inhibitor).

d- Calculation of the erosion velocity by weight loss method:

In term of accurate analytical scale, each experiment was repeated thrice, the deviated results were excluded, and the experiments were repeated. The represented results are the medium of the thrice acceptable measurements.

The erosion velocity was calculated according to the following relationship:

$$V_{Corr} = \frac{m}{S \ . \ t}$$

V_{Corr}: Velocity of corrosion; Δm : mass variation (g); S: sample area (cm^2); t: immersion time (min).

The determination of the surfaces of the simples were realised from measuring their dimensions in cm.

e- Inhibition yield:

The inhibitory effectiveness (I_{Corr} %) is given by the following relation:

$$I_{Corr}\ \% = V_0 - \frac{Vinh}{V_0}\ x\ 100$$

V_0: corrosion velocity in the absence of the inhibitor.

V_{inh}: corrosion speed in the presence of the damper.

3. Results and Discussion

The weight loss method remains in use today for studying the inhibition of metal corrosion due to its simplicity, convenience, and the absence of a requirement for expensive equipment. Herein, we employ this method to conduct our experimental research. The measurements were conducted both in the absence and presence of the inhibitor. We considered three parameters: immersion time, experiment temperature and inhibitor concentration in the corrosive medium were considered.

The first set of measurements involves determining the impact of the acidic medium on the carbon steel X70 in the absence of the inhibitor, with varying immersion duration.

a- Effect of immersion time in corrosive medium on the X70 steel

The chosen durations of the immersion were 30, 60, 90, 120, 150 and 180 minutes. Experimental values of steel X70 mass loss as a function of time in acidic medium (0.5 M H_2SO_4) in the absence of the inhibitor at T = 25 °C are grouped in table 1.

Table 1: Elemental percentages of the XC70 steel used m_0 = 21.2985 g.

t (min)	0	30	60	90	120	150	180
m_1 *(g)*	21.2985	21.2921	21.2886	21.2837	21.2823	21.2797	21.2772
$\Delta m = m_0 - m_1$ *(g)*	0.0000	0.0051	0.0099	0.0134	0.0162	0.0188	0.0213

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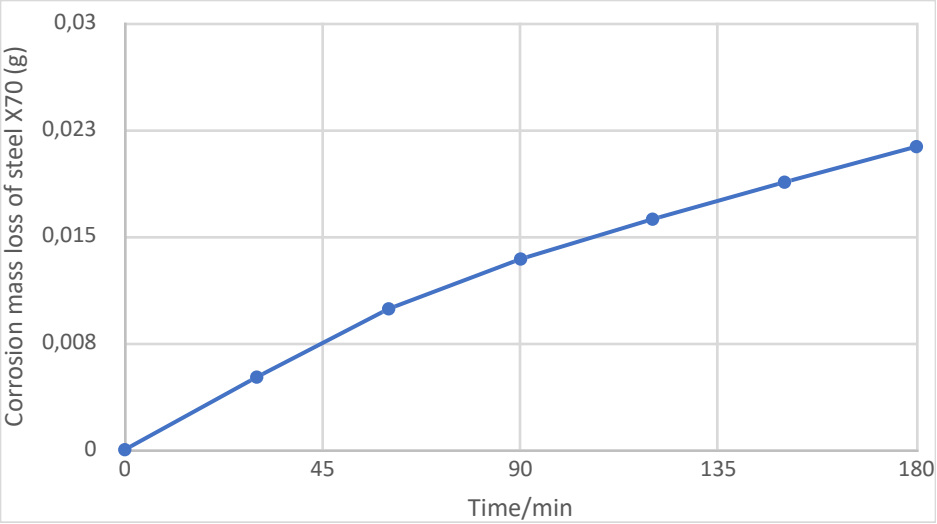

Figure 2. Mass loss of steel X70 (m_0 = 21.2985 g) as a function of time in acidic media, (0.5 M H_2SO_4) in the absence of the inhibitor at T = 25 °C.

From the results of table 1, traduced into the graphic of figure 2, the immersion duration was found to be among the factors that have a high impact on the corrosion rate of the carbon steel.

On the other hand, and as grouped in table 2, and traduced in the graphic of figure 3, the calculated values of the velocity decreases of the corrosion with the increase of the sample immersion duration for the same corrosion measurements. This decrease comes from the spontaneous formation of the steel *anti*-corrosion protective layer, which reduces its dissolution.

Table 2. Corrosion speed and time of steel immersion in (0.5 M H_2SO_4) at 25 °C.

t (min)	30	60	90	120	150	180
$V \cdot 10^{-5}$ $(g.cm^{-2}.min^{-1})$	1.293	1.131	0.983	0.862	0.760	0.671

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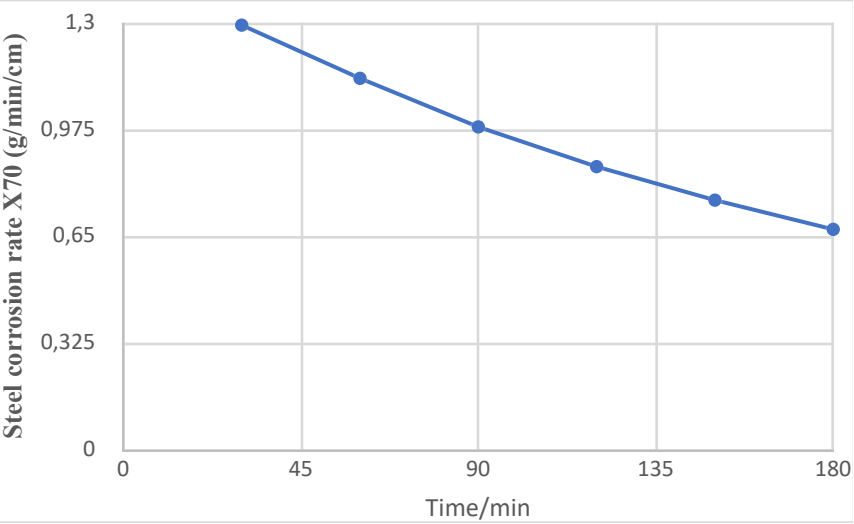

Figure 3. The decrease of the rate of corrosion of the steel X70 in terms of time in an acidic medium (H_2SO_4 0.5 M in the absence of an inhibitor).

b- Study of the speed and effectiveness of corrosion in the presence of the Nutmeg Oil

In this part of the study, we calculate the corrosion speed, the inhibitory efficiency (% I_{Corr}) as and the rate of steel corrosion velocity after immersion in the absence and the presence of the inhibitor at different concentrations at room temperature, the results are shown in table 3.

Table 3. Corrosion rate and inhibition effectiveness, in the presence and absence of the inhibitor.

Entry	*C ($g.L^{-1}$)*	*m_1 (g)*	*m_2 (g)*	*S (cm^2)*	*(%) I_{corr}*	*$V.10^{-5}$ ($g.cm^{-2}.min^{-1}$)*
1	0.0625	17.7990	17.7785	10.98	42.13	1.585
2	0.1250	17.4311	17.4226	10.62	49.09	1.374
3	0.2500	19.8646	19.8545	10.62	57.13	1.136
4	0.5000	18.5950	18.5873	10.68	60.65	0.780
5	1.000	19.9008	19.8925	11.30	64.69	0.240

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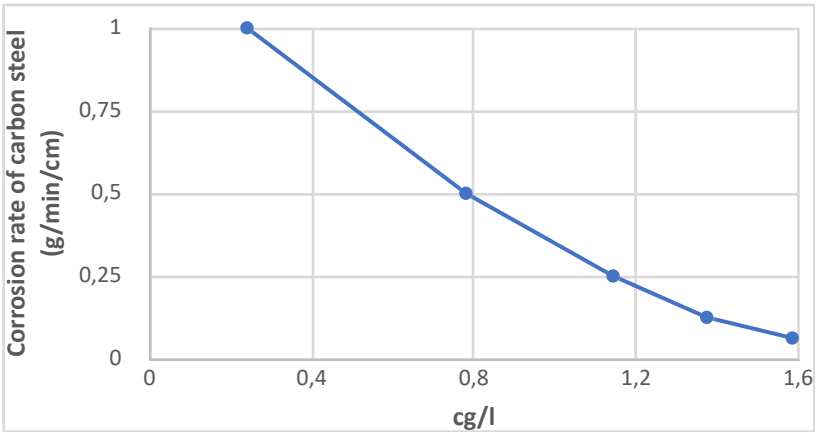

Figure 4: Corrosion speed change in terms of concentration

As shown in table 3, the extract has good properties to prevent corrosion of steel. Through these results we concluded that the ideal concentration for the highest inhibition activity is 1 $g.L^{-1}$, with a ratio of 64.69 %, and we recorded at this concentration the lowest corrosion speed is 0.240 $g.cm^{-2}.min^{-1}$)

c- The effect of temperature in the presence of the inhibitor

Based on the previous results, we consider that an hour of immersion is a sufficient during to study the effect of the temperature on the inhibition process in corrosive medium. So, in the inhibition prepared solutions were experimented at different temperatures 30.5, 50.2 °C and 70 °C, to measure the mass loss to study the impact of temperature on the phenomena.

From the experimental results grouped in table 4, we conclude that the temperature has a substantial impact on the efficiency of the inhibition, with the highest inhibition rate of 86.25 % recorded at 30.8 ^{0}C and the lowest inhibition rate of 14 % recorded at 70.0 °C, for the lowest inhibitor solution concentration, indicating that the inhibitor's action was hampered by this temperature. We find that an increase of the concentration of the inhibitor solute, increases its efficiency as inhibitor. Whereas an increase of the temperature decreases its inhibition efficiency. That better inhibition results were observed at 30.8 °C for a concentration inhibition solution of 1.000 $g.L^{-1}$, while the poor one corresponds to the lowest concentration inhibition value (0.125 $g.L^{-1}$) at 70 °C.

Table 4. The effect of temperature on the inhibitory activity of the oil.

T (^{0}C)	30.8 (^{0}C)	50.2 (^{0}C)	70.0 (^{0}C)
C_{inh} (g/L)	I %	I %	I %
0.125	35.19	18.26	14.00
0.250	41.38	21.57	18.82
0.500	51.18	45.55	26.27
1.000	86.25	58.48	39.03

Conclusion

Plant extracts of natural origin are available in various forms and represent an excellent option to replace toxic, harmful and expensive synthetic corrosion inhibitors. In addition to being optionally certified, inexpensive, biodegradable, and non-toxic, these green inhibitors possess a variety of phytochemicals that can be present on the surface of the substrate and form a shield.

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References

[1] S. Z. Salleh, A. H. Yusoff, S. K. Zakaria, M. A. A. Taib, A. Abu Seman, M. N. Masri, M. Mohamad, S. Mamat, S. Ahmad Sobri, A. Ali, P. Ter Teo., Plant extracts as green corrosion inhibitor for ferrous metal alloys: a review, *J. Clean. Prod.*, **2021**, *304*, 127030.

[2] S. A. Umoren, M. M. Solomon, I. B. Obot, R. K. Suleiman, A critical review on the recent studies on plant biomaterials as corrosion inhibitors for industrial metals, *J. Ind. Eng. Chem.*, **2019**, *76*, 91-115.

[3] A. Marciales, T. Haile, B. Ahvazi, T. D. Ngo, J. Wolodko, Performance of green corrosion inhibitors from biomass in acidic media, *Corros. Rev.*, **2018**, *36*, 239-266.

[4] E. Alibakhshi, M. Ramezanzadeh, S. A. Haddadi, G. Bahlakeh, B. Ramezanzadeh, M. Mahdavian, Persian Liquorice extract as a highly efficient sustainable corrosion inhibitor for mild steel in sodium chloride solution, *J. Clean. Prod.*, **2019**, *210*, 660-672.

[5] S.-H. Yoo, Y.-W. Kim, K. Chung, S.-Y. Baik, J.-S. Kim, Synthesis and corrosion inhibition behaviour of imidazoline derivatives based on vegetable oil, *Corros. Sci.*, **2012**, *59*, 42-54.

[6] panel A. Dehghani, G. Bahlakeh, B. Ramezanzadeh, M. Ramezanzadeh, Potential role of a novel green eco-friendly inhibitor of mild steel in HCl solution: detailed macro/ micro-scale experimental and computational explorations, *Constr. Build. Mater.*, **2020**, *245*, 118464.

[7] A. Dehghani, G. Bahlakeh, B. Ramezanzadeh, M. Ramezanzadeh, Aloysia citrodora leaves extract corrosion retardation effect on mild-steel in acidic solution: molecular/ atomic scales and electrochemical explorations, *J. Mol. Liq.*, **2020**, *310*, 113221.

[8] lumo, O. J. Odejobi, E. L. Odekanle, Thermodynamics and adsorption study of the corrosion inhibition of mild steel by Euphorbia heterophylla L. extract in 1.5 M HCl, *Results Mayer.*, **2020**, *5*, 100074.

[9] A. Dehghani, G. Bahlakeh, B. Ramezanzadeh, M. Ramezanzadeh, Potential of Borage flower aqueous extract as an environmentally sustainable corrosion inhibitor for acid corrosion of mild steel: electrochemical and theoretical studies, *J. Mol. Liq.*, **2019**, *277* 895-911.

[10] M. Mobin, M. Basik, J. Aslam, Pineapple stem extract (Bromelain) as an environmental friendly novel corrosion inhibitor for low carbon steel in 1 M HCl, *Measurement*, **2019**, *134*, 595-605.

[11] V. C. Anadebe O. D., Onukwuli, M. Omotioma, N. A. Okafor, Experimental , theoretical modelling and optimization of inhibition efficiency of pigeon pea leaf extract as anti-corrosion agent of mild steel in acid environment, *Mater. Chem. Phys.*, **2019**, *233*, 120-132.

[12] M. Abdallah, H. M. Altass, A. S. Al-Gorair, J. H. Al-Fahemi, B. A. A. L. Jahdaly, K. A. Soliman, Natural nutmeg oil as a green corrosion inhibitor for carbon steel in 1.0 M HCl solution: chemical, electrochemical, and computational methods, *J. Mol. Liq.*, **2021**, *323*, 115036.

[13] R. Haldhar, D. Prasad, Le T. D. Nguyen, S. Kaya, I. Bahadur, O. Dagdag, S.-C. Kim, Corrosion inhibition, surface adsorption and computational studies of Swertia chirata extract: a sustainable and green approach, *Mater. Chem. Phys.*, **2021**, *267*, 124613.

[14] Q. Xiang, J. He, Combining theoretical and experimental researches to insight the *anti*-corrosion nature of Citrus reticulata leaves extract, *J. Mol. Liq.*, **2021**, *325*, 115218.

[15] H. Li, Y. Qiang, W. Zhao, S. Zhang, A green Brassica oleracea L extract as a novel corrosion inhibitor for Q235 steel in two typical acid media, *Colloids Surf. A Physicochem. Eng. Asp*, **2021**, *616*, 126077.

[16] R. Haldhar, D. Prasad, I. Bahadur, O. Dagdag, A. Berisha, Evaluation of Gloriosa superba seeds extract as corrosion inhibition for low carbon steel in sulfuric acidic medium: a combined experimental and computational studies, *J. Mol. Liq.*, **2021**, *323*, 114958.

[17] M. H. Shahini, M. Ramezanzadeh, G. Bahlakeh, B. Ramezanzadeh, Superior inhibition action of the Mish Gush (MG) leaves extract toward mild steel corrosion in HCl solution: theoretical and electrochemical studies, *J. Mol. Liq.*, **2021**, *332*, 115876.

[18] K. Krishnaveni, J. Ravichandran, Influence of aqueous extract of leaves of Morinda tinctoria on copper corrosion in HCl medium, *J. Electroanal. Chem.*, **2014**, *735*, 24-31.

[19] R. Laggoun, M. Ferhat, B. Saidat, A. Benghia, A. Chaabani, Effect of *p*-toluenesulfonyl hydrazide on copper corrosion in hydrochloric acid solution, *Corros. Sci.*, **2020**, *165*, 108363.

[20] L. Chen, D. Lu, Y. Zhang, Organic Compounds as Corrosion Inhibitors for Carbon Steel in HCl Solution: A Comprehensive Review. *Materials (Basel)*, **2022**, *15(6)*, 2023.

[21] A. O. Errayes, G. A. Alshibani, M. O. Darwish, Corrosion Inhibition of carbon steel XC70 in two acidic media (0.1 N H_2SO_4 and 0.1 N HCl) by weight loss method using Black Pepper Extract as corrosion inhibitor, *Libyan J. Basic Sci.*, **2023**, *21(1)*, 75-89.

[22] B.D. Manuel, S. Marsela, Trimyristin isolation from nutmeg and synthesis of methylester using heterogeny catalyst, *Ind. J. Chem Res.*, **2014**, 2, 60-165.