



The Inhabitation Performance of Tarmaix Material Upon Mild Steel Substrates in an Acid Medium

Worda Shaha^{1*}, Mousa May¹, Khadija Khalifa¹, Balhassn Ali²

¹Chemical Engineering Department, Faculty of Engineering, Sebha University, Sebha, Libya

²College of Petroleum Engineering, Al-Jafra University, Zalla, Libya

Email: *mou.may@sebhau.edu.ly

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Abstract

Metallic materials are used in many applications due to their mechanical and physical properties. Under an aggressive environment, *i.e.* high temperature and/or corrosive environment these materials can be degraded at a fast rate. One way to protect the metal surface against corrosion is to apply the organic inhibitor material. The inhibitor will normally be chemically adsorbed by the surface of the metal; this will form a protective thin film. The current work is to investigate the influences of the Tarmarix plant as an organic material source for inhibiting carbon steel in corrosion media. The obtained results show that the changes in weight loss because of the corrosion in samples immersed in corrosion media containing 10% and 20% of Tarmaix; were less than in samples immersed in the same media without the Tarmaix. The corrosion rate decreased with time for all inhabitation specimens. The results revealed that longer time exposure to the inhibitor material results in higher inhibition efficiency which was above 90% efficiency. And the plots of $C\theta$ versus C give straight lines with the linear correlation coefficient (R^2) values close to unity, which follow the Langmuir adsorption isotherm. The slope ranges of these lines are 1.391 - 1.623 within the used immersion times. Therefore, it can be suggested that the absorbed molecules form a monolayer on the mild steel surface. The strong appearance of functional groups such as C-OH inside the structure of polyphenolics in the plant leaf extract of Tarmaix could probably be responsible for the reduction of metal ions and the formation of a protective layer that improved the corrosion process within the inhabitation samples.

Subject Areas

Composite Material

Keywords

Mild Steel, Tarmaix Plant, Corrosion, An Organic Inhibitor

1. Introduction

Metallic material is widely used in many engineering applications, including applications that involve working under high temperatures and/or stress levels, such as steel components used in power plants, chemical plants, and oil refineries. The metallic materials have many good qualities including keeping most of their mechanical properties unchanged even when working under a wide range of temperatures. However, corrosion in metallic materials is one of the main issues facing design engineers when designing components working in a corrosive environment. Steel is one of the main metals used to make engineering components, however, it has the tendency to react with it is surrounding forming an unstable protection layer. In the past few years, organic materials have been used widely to reduce corrosion of metals, because of their efficiency, low cost, and low toxicity.

Many efforts have been made, and attempts are still being made to make the steels immune or passive towards their surrounding environment [1]. These include developing and utilizing suitable inhibitors for corrosive environments or new types of paints or surface coatings. It is well-known fact that organic materials generally have hetero atoms, such as O, N, and S [2]. The use of organic compounds containing oxygen, sulfur, and nitrogen elements to reduce corrosion attacks of steel has been deeply investigated elsewhere. Generally, those compounds have a great ability to be adsorbed on the surface of mild steel, leading to blocking the active corrosion positions. However, there are several problems related to chemical inhibitors, such as their high cost and the integrative effect on the environment. Tarmaix (athel tamarisk) is a shrub or a tree found along streams, and lake shores and is still widely planted in desert regions [3]. Nowadays, extracts of natural substances have been used due to their potential as eco-friendly, non-toxic, and good corrosion inhibitors.

It was stated [4] that the role of inhibitors is to form a barrier of one or several molecular layers against acid attack on the metal surface. Chemical and physical adsorption processes are often associated with involving a variation in the charge of the adsorbed substance and transfer of charge from one phase to the other. The inhibitor compounds have found useful applications in the formulation of primers and anticorrosive coatings. The alternative corrosion inhibitors, organic substances containing polar functions with nitrogen, sulfur, and oxygen in the conjugated system have been reported to exhibit very good inhibiting properties on metallic substrates [5]. The inhibitive characteristics of such compounds derive from the adsorption ability of their molecules, with the polar group acting as the reaction center for the adsorption. The resulting adsorbed film acts as a barrier that separates the metal from the corrosion, and the efficiency of inhibition

depends on the mechanical, structural, and chemical characteristics of the adsorption layers formed under particular conditions [6] [7]. Inhibitors are often added in industrial processes to secure metal dissolution from acid solutions. Due to a lack of information on using Tarmaix as an organic inhibitor to protect metals in acid media. This work will examine the use of the extract of Tarmaix collected from the area in the south of Libya (Sebha Province), as a corrosion inhibitor for carbon steel immersed in an acidic medium.

2. Experimental Procedures

2.1. Materials

Carbon steel specimens with dimensions of $29 \times 29 \times 0.2$ cm have been used in this work. In all measurements, samples were immersed in corrosion media for a whole surface area. A small cotton wire has been used to keep specimens within the solution.

2.2. Preparation of the Plant Extract

Tarmaix plant is used in this work as an organic material source to inhibit carbon steel in corrosion media. The Tarmaix slender branches were purchased from a local market, in Sabha, Libya. For extraction, the branches of Tarmaix slender were extracted with acetone for 6 hrs, and then have been washed and air dry and grind fine, and then shifted into powder form. For preparing 10% conc. 10 g of Tarmaix powder was rinsed in 100 ml deionized water and then placed and kept in an oven at 80°C for 2 hrs. The solution was filtered in a water bath until the amount reached 10 ml. The concentrated solution was then removed and left to cool to room temperature. For the preparation of 20, 30, 40, and 50 different percentages of Tarmaix inhibitors, the same procedure was applied. A specified amount of Tarmarix inhibitor was directly added to the corrosive medium. **Figure 1** shows (a) Tarmarix powder, (b) acetone and (c) deionized water used in the experiments.



Figure 1. Tamarix, acetone and deionized water used.

The aggressive media used in this experiment was made of 37% HCl, which is an appropriate concentration of acid and this concentration was the same for all specimens. To prepare the inhibitor, a sensitive balance is used to measure the weights of the chemicals used in the experiment. A drying oven was used to dry samples and a water bath was used to incubate samples in water at a constant temperature over a long period of time. Also, a filter paper or semi-permeable paper is used to separate the fine solids in a liquid during a filtration process. A sample preservation tube is used to keep samples from mixing with other materials or external factors such as temperature.

2.3. Corrosion Rate Measurements

Weight loss measurements of C-steel specimens were abraded with emery papers of 1/0, 2/0, 3/0, and 4/0 grade, degreased by acetone and weighed, then immersed in 100 ml of the test solution in open beakers. Steel specimens were immersed in 100 mL of electrolyte with and without the Tarmarix solutions for 1, 2, 3, 4 and 5 hrs at room temperature. The cleaned specimens were weighed before and after immersion into the corrosive solution. The difference in weight after each interval was taken as weight loss. The experiment was repeated 3 - 5 times. The tested specimens were removed, rinsed with water and acetone, and dried. The loss of weight of the steel specimen was determined using a digital balance with the precision of $0.0001 \pm g$ for each 0.1 mg. The weight loss against the inhibitor concentration was plotted in **Table 1**. The inhibition efficiency (% IE) of the inhibitor, degree of surface coverage (θ), and corrosion rate (CR) was made from the reading recorded in **Tables 2-3** using the following equations.

$$\text{Inhibition Efficiency: \% IE} = (1 - W_1/W_2) \times 100 \quad (1)$$

where W_1 and W_2 are the weight losses for specimens in the presence and absence of the Tarmarix plant inhibitor used.

$$\text{Surface Coverage: } (\theta) = (CRB - CRW)/CRB \quad (2)$$

where CRB and CRW are the corrosion rates in the absence and presence of the inhibitor

$$\text{Corrosion Rate: CR (mmy}^{-1}) = 87.6W/At \quad (3)$$

where W is the weight loss, (A) is the exposed specimen area (cm^2), (t) is the exposure time (hr) and (d) is the specimen density (g/cm).

2.4. Fourier Transform Infrared (FTIR)

It is well known that the FTIR offers quantitative and qualitative analysis for organic and inorganic samples. Fourier Transform Infrared Spectroscopy (FTIR) identifies chemical bonds in a molecule by producing the absorption spectrum. A Nicolet Nexus FTIR spectro-photometer from scientific research and consulting center at Sebha University was used in this work to measure, identify and detect functional groups and to characterize covalent bonding information within the material.

3. Result and Discussions

3.1 Weight Loss Measurements

In this work, weight loss evaluation was used to assess the efficiency of the inhibition of Tarmarix plant in hydrochloric acid media on mild steel corrosion. Different Tarmarix (plant extracts) concentrations were used at room temperature in a corrosive medium in order to examine its effect on the inhibition process of mild steel. There are three variables in this test (i) the existence of the Inhibitor, (ii) Inhibitor concentration and (iii) immersion time. The efficiency of the Inhibitor was measured using the loss of weight theory, these test results are reported in **Table 1**. The corrosion rate in millimeters per year (mm/yr) was determined by Equation (3).

Table 1. The effects of the Immersion time (h) on samples weight (gr).

Immersion Time, Hrs	Inhibitor Concentration, wt%					
	0%	10%	20%	30%	40%	50%
Original weight, gr	17.2317 gr	17.6747 gr	27.2904 gr	28.9370 gr	22.2771 gr	23.3067 gr
1 hrs	16.8701 gr	17.0701 gr	27.0335 gr	28.5760 gr	22.1317 gr	23.2036 gr
2 hrs	15.1375 gr	16.9982 gr	26.8511 gr	28.4259 gr	22.0283 gr	23.1053 gr
3 hrs	12.7403 gr	16.7519 gr	26.5657 gr	28.1906 gr	21.8379 gr	22.9377 gr
4 hrs	11.7751 gr	16.5679 gr	26.2423 gr	27.8482 gr	21.5583 gr	22.7160 gr
5 hrs	11.2549 gr	16.2915 gr	26.0327 gr	27.5841 gr	21.2761 gr	22.487 gr

The results presented in **Figure 2** show the effects of using Tarmarix material as an organic inhibitor against the corrosion process of mild steel in an acid media. The concentrations of inhibitors in this test vary between 0% to 50%. It was noted that organic additives prevent the adsorption of chloride ions on the surface and/or may help the formation of a resistant oxide layer on the metal surface [8]. In addition, the results indicated slight changes in weight losses as immersion time increased (from 1 hr up to 5 hrs). The reason might be related to more resistance being achieved as Tarmarix concentration increased. This is explained in term of the presence more organic chemical crosslink structure in corrosion media led to multi chemical bonds playing a barrier for reaching corrosion ions into the metal surface. Also, it was mentioned that a number of factors including the nature of metal, testing media, chemical structure of inhibitor, and nature of substituents present in the inhibitor influence the nature of inhibitors adsorption [9] [10]. The containing nitrogen, oxygen, and sulfur atoms in organic compounds and having multiple bonds will improve the inhibition process for the corrosion of many metals and alloys [11]. The result shows that the increase in the concentration of the inhibitor led to a reduction in weight loss. This attribute to the Tarmarix material plays a role in reducing the attaché of chlorine ions upon the substrate surface [12] [13].

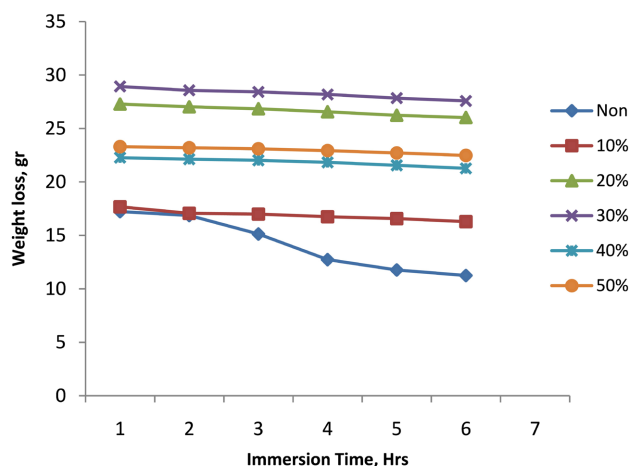


Figure 2. Changes in weight over-time for samples with and without inhibitors.

3.2. Corrosion Rate Calculation

The corrosion rate for each concentration level of the inhibitors over multiple time spans are provided. It can be seen from the results presented in **Table 2**, that the corrosion rate decrease as the inhibitor concentration increases despite the exposure time. The corrosion rate has increased significantly over time for specimens emerged in the acidic liquid without Tarmaix inhibitor. In addition, the Tarmaix material has a good deposit on the electrode surface and causes passivation on the corroded electrode which led to becoming less affected by its environment [14]. This behavior was supported by much research, which reported [15] [16] that plant extracts contain natural compounds such as tannins, alkaloids, flavonoids, phenols, and organic acids that have been found to possess inhibitive properties against corrosion. The active compounds in these materials can form a protective film on the metal surface, which acts as a barrier against corrosive agents.

Table 2. Corrosion rate using different concentration level of the inhibitors over multiple time spans.

Time, Hrs	Corrosion rate, mm/yr					
	Non	10%	20%	30%	40%	50%
1	0.1681	0.2811	0.1194	0.1678	0.0676	0.0474
2	0.4868	0.1572	0.1021	0.1188	0.0578	0.0468
3	0.5538	0.1430	0.1123	0.1160	0.0680	0.0371
4	0.6142	0.1286	0.1286	0.1265	0.0635	0.0356
5	0.6355	0.1288	0.1169	0.1260	0.0623	0.0331

3.3. Inhibitor Efficiency

This section discusses the changes in the inhibition efficiency overtime in the corrosive medium. The results revealed that the inhibitor with 50 % concentration after 5 hrs has a higher inhibition efficiency which is about 90% see **Figure 3**. The comparison results in **Figure 2**, also indicate that the presence of Tarmaix material

as an organic additive within the solution reduces the corrosion rate in all inhibitor concentrations compared with samples tested without an inhibitor. Furthermore, the increase in Inhibitor efficiency is a result of the increase in the adsorption rate of the inhibitor within the solution which leads to a complete blockage of corrosion; this assumption was supported by other research in [17] [18].

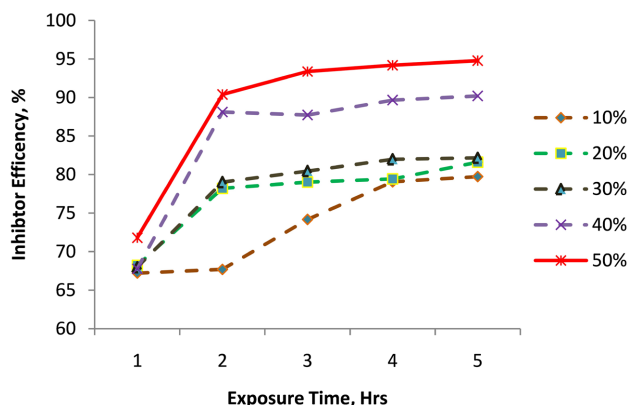


Figure 3. Shows the changes in inhibitor efficiency over time.

3.4. Adsorption Study

The adsorption study applies to determine the way by which the inhibitor molecules interact with the metal substrate surface. The values of surface coverage, θ , for different Tarmaix inhibitor concentrations versus immersion times were exhibited in Figure 4. It is well known that [19] Langmuir isotherms of adsorption is one model used to determine the adsorption ability of inhibitor on metal surface. In addition, the surface coverage was studied graphically by suitable adsorption models. The plots of C/θ versus C give straight lines with the linear correlation coefficient (R^2) values close to unity, which follow the Langmuir adsorption isotherm. The slope ranges of these lines are 1.391 - 1.623 within the used immersion times, as shown in Figure 3. Therefore, it can be suggested that the absorbed molecules form monolayer on mild steel surface. This was supported by other results in [20]-[22] on corrosion inhibition of mild steel in HCl solution.

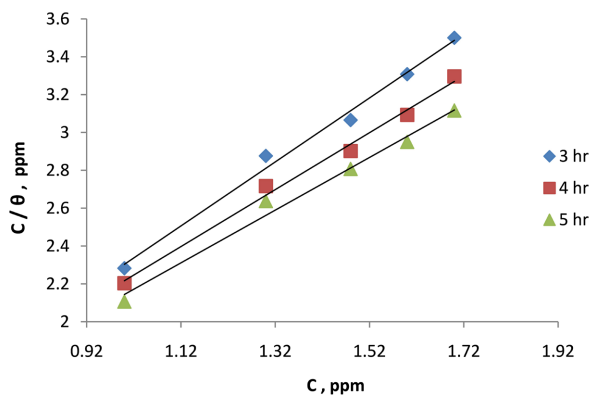


Figure 4. Longmuir's adsorption isotherm plots for the adsorption of Tarmaix in HCl on mild steel at different immersion times.

3.5. IR Spectral Studies

The FTIR spectra were recorded to understand and investigate the interaction of inhibitor molecules with the metal surface. It can be seen that the spectra depicted some peaks at different wavenumbers which represent free OH in molecules and OH group forming hydrogen bonds, hydrocarbons, carbonyl group (C=O), stretching C=C aromatic ring and C-OH stretching vibrations within the structure (See **Table 3**).

Table 3. Chemical bonds and wavenumbers.

Chemical bonds	Wavenumbers, cm^{-1}
C=O	2800 - 3000
-C=O	1650 - 1850
-OH or -NH	3000 - 3500
C-C	1400 - 1500
-CH	2800 - 3000
C=N	1050 - 1260
C-OH	1000 - 1300

Figure 5(A) represents the IR spectra of (metal surface + HCl) system. The strong appearing of functional groups such as C-OH (**Figure 5(B)**) inside the structure of polyphenolics in the plant leaf extract of Tarmaix could be probably responsible for the reduction of metal ions and formation of a protective layer improved the corrosion process within the inhabitation samples [23]. Moreover, very strong sharp of carboxyl group appeared in the range of $3000 - 3500\text{cm}^{-1}$, in **Figure 5(B)** compared with that in **Figure 5(A)**. This may be attributed to the presence of oxides metals within the Tarmaix structure which help to improve the corrosion resistance of metal surface within the solution.

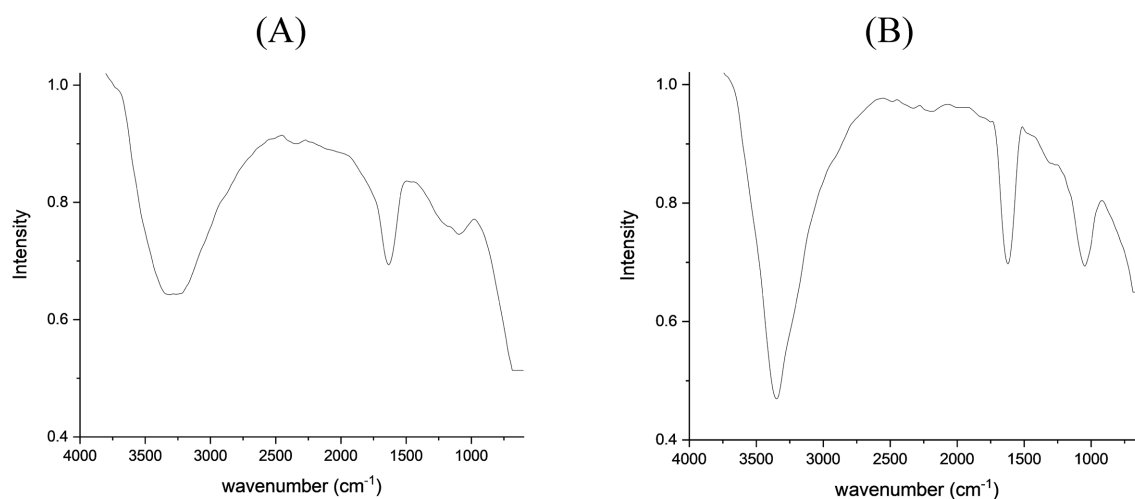


Figure 5. IR spectra of (A) without inhibitor and (B) with Tarmaix inhibitor.

4. Conclusions

1) It was observed that natural plant extracts are very effective green corrosion inhibitors for mild steel.

2) The weight loss was less in samples immersed in corrosion media with high concentrations of Tarmaix material, also the loss of weight was significantly high for specimens tested without the presence of the Tarmaix material.

3) The specimen's corrosion rate decreases with exposure time for all tested inhabitation concentrations (from 0% to 50%). Also, after 5 hours of exposure and 50% Tarmaix concentration, the inhibitor material showed a higher inhibition efficiency which was slightly above 90%.

4) The plots of C/θ versus C give straight lines with the linear correlation coefficient (R^2) values close to unity, which follow the Langmuir adsorption isotherm.

5) The strong appearance of functional groups such as C-OH (**Figure 5(B)**) inside the structure of polyphenolics in the plant leaf extract of Tarmaix could probably be responsible for the reduction of metal ions and formation of a protective layer improved the corrosion process within the inhabitation samples.

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Conflicts of Interest

The authors declare no conflicts of interest.

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