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Corrosion Inhibition of Mild Steel in Acidic Medium Using Ethanolic Extract of Christ Thorn Leaves

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Abstract

The study involves the corrossion inhibition of mild steel in acidic medium using ethanolic extract of christ thorn leaves. An experimental extract of Christ thorn leaves was carried out in 0.9 M HCl solutions using the weight loss technique. The result has proved that the extract is a good inhibitor of corrosion of mild steel in HCl. The inhibition efficiencies ranged from 47.15% to 60.04% and 44.31% to 54.09% at 303 and 323 K respectively. The corrosion rate C_R *decreases with an increase in the concentration of inhibitors. Moreover, surface coverage (θ) and percentage inhibitor efficiency (% IE) increase with increasing concentrations of inhibitor. The adsorption of the inhibitor on the surface of mild steel was found to be exothermic, spontaneous and consistent with the mechanism of physical adsorption as the value for heat of adsorption ranged from -22.04 to -17.73* $kJmol^{-1}$*. The adsorption data fitted well to the Langmuir adsorption isotherm. The surfaces morphology also revealed that severe corrosion on the metal surfaces in the uninhibited solution, indicating uniform damage characterized by cracks across the entire metal surface, but by the addition of the plant extracts the metal surfaces were effectively inhibited.*

Keywords: Adsorption, Christ's thorn, Inhibition, Mild Steel, SEM.

INTRODUCTION

The term corrosion also describes the disintegration or degradation of materials brought on by an environment-based chemical or electrochemical reaction (Amitha and Bharathi, 2011). Corrosion is a naturally occurring phenomenon generally defined as the deterioration of a substance (usually a metal) or its properties when exposed to an aggressive environment through a chemical or electrochemical reaction (Karthik *et al.*, 2015). Metal deterioration due to chemical attack or reactivity with surroundings also referred to as corrosion. It is an ongoing and persistent problem that is often difficult to completely eliminate but it can be prevented (Nishimura *et al.*, 2012).Corrosion processes involve a number of reactions that intensify when the protective barrier is broken, including oxide formation, electrochemical potential, migration of metal cations into the coating matrix, and local pH changes. An enormous amount of interest has been shown since iron and alloys corrosion is a topic of significant theoretical and practical importance (Karthik *et al.,* 2015).

Raw resources derived from plants are inexpensive, readily available, and sustainable. Their extracts which are composed of organic compounds with atoms of nitrogen, sulfur, and oxygen come from their roots, fruits, seeds, barks, and leaves. These extracts have been shown to be efficient inhibitors (Mohammad *et al.,* 2020). Preliminary phytochemical screening of the ethanolic extracts of Christ thorn in our laboratory and report by other authors (Kolo *et al.,* 2016) have revealed that the extracts is a complex mixture of various phytochemical components such as saponins, flavanoids, tannis, alkaloids, organic acid are present in the leaves but only anthraquinones is absent in the extracts. The primary adsorption centers of these organic compounds are triple or conjugated double bonds or aromatic rings, which are found in their molecular structures and typically feature polar functionalities with nitrogen, sulfur, or oxygen atoms (Abdallahi *et al.,* 2014).

Recent developments in the knowledge of the mechanism and phenomena of corrosion have been introduced to address them; they led to the development of materials with higher corrosion resistance, more appropriate prevention and corrosion control method (Mas *et al.,* 2020). A substance that lowers the rate of corrosion in an environment when added in modest amounts is called an inhibitor. Compounds with hetro-atoms like oxygen, nitrogen, phosphorus and sulfur that permit adsorption on a metallic surface are the most effective and competitive natural inhibitors (Umoren *et al.,* 2011).

Corrosion inhibitors are typically costly, synthetic substances that have the potential to taint the final product's quality. The organic compounds are hazardous to the environment and harmful, there is growing concern about non-toxic, affordable, and suitable inhibitors (Joseph *et al.,* 2012). Consequently, corrosion scientists are interested in researching sustainable corrosion inhibitors. Biodegradable, non-toxic, environmentally safe, and less expensive are the main characteristics of inhibitors derived from natural sources (Asgarpanah and Haghighat, 2012).

MATERIALS AND METHODS

Phytochemical screening

Chemical tests were carried out on the ethanolic extract of Christ's thorn and on the powdered extracts using standard procedures as described by Sofowara (1993), Trease and Evans (1989) and Harborne (1973) to identify the following phytochemical constituents such as Test for tannins, saponins, alkaloids, flavonoids, terpenoids, carbohydrates, anthraquinones, steroid/triterpenes and cardia glycosides (Anindita and Bikramjit, 2017).

Preparation of Plant Extract

The aggressive test medium of 0.9 M HCl acid stock solutions were prepared using analytical-grade 37% of HCl with distilled water. The concentrations of the plant extract were ranged from 0.2-0.8 g/L for this study. These procedures were as described by Mohammed *et al.,* (2020).

Weight Loss Measurements

The mild steel (MS) coupons were investigated by optical emission spectroscopy (OES) with the following composition (wt%): 0.15% C, 0.03% Si, 0.6% Mn, 0.36% P, 0.36% S, and 98.5% Fe. After polishing the sheet using emery papers ranging in grade from 400 to 1200, it was made into dimensions of (2.5 x 2.5 x 0.4 cm) in length, width, and thickness respectively. It was then cleaned with ethanol, rinsed with acetone, and allowed to dry at room temperature. In this investigation, distilled water was utilized along with all analytical-grade reagents (Al-Otaibi *et al.,* 2014). The temperatures for each run of this study were kept constant using a digital thermostatic water bath. In this procedure, nine 250 mL beakers were used and properly labeled. One hundred mill (100 mL) each of the prepared Christ thorn leaves stock solution (that is 0.2, 0.4, 0.6 and 0.8 g/L) were transferred into the first set of eight beakers. Into the nineth beaker, 100 ml of each uninhibited stock solution 0.9M HCl (that is control or blank) were transferred. All the beakers containing the test solutions were placed into the thermostated water bath. After the set-temperature attained a steady state, the clean, weighed, tag coupons from the desiccator were introduced and completely immersed into the test solutions in the water bath and suspended with the aid of pegs via the embroidery thread. After 1 hour, the test solution's coupons were removed, and then put back into the solutions after being cleaned, dried, and weighed. The processes were repeated for a period of 4 hours and the differences in the weight loss for this period were taken as total weight loss.

Equation (1) was used to compute the total weight reduction.

$$\mathrm{W_i} - \mathrm{W_f} = \mathrm{W} ------ \qquad (1)$$

Where $\mathrm{W_i}$and $\mathrm{W_f}$ are the starting weight is before immersion and where it is after, respectively. Equation (2) provides the degree of surface covering(θ).

$$\theta = \left(1 - \frac{\mathrm{w_1}}{\mathrm{w_2}}\right) --------- (2)$$

Where $w_1$and $w_2$are the metal weight losses (g) in a 0.9 M HCl solution when the inhibitor is present and absent.

Equation (3) was used to get the inhibition efficiency (%IE) for each inhibitor concentration.

$$\%\mathrm{IE} = \left(1 - \frac{\mathrm{w_1}}{\mathrm{w_2}}\right) \times 100 ----- \qquad (3) \quad \text{(Onen, 2016)}$$

Equation (4) was used to calculate the metal corrosion rates in the acid concentration over a 4-hours immersion time starting from weigh loss.

$$\mathrm{CR(mmpy)} = \frac{87.6\mathrm{w}}{\mathrm{DAT}} ----- (4)$$

Where D is the specimen's density (g/cm3), A is the specimen's area (square meters), T is the immersion time (hours), w = Total weight loss (mg) and 87.6 is the conversion factor.

Thermodynamic/Kinetic Parameters

By plotting the natural logarithm of the corrosion rate (log C_R) versus 1/T both with and without the addition of different inhibitor doses, the activation Energy Ea for the inhibited corrosion reaction of the metals was visually estimated from the slope using the Arrhenius equation (5).

$$CR = Ae^{\frac{-E_a}{RT}} ------- \qquad (5)$$

Equation (6) was obtained when we take logarithms of both side of equation (5)

$$logCR = -\frac{E_a}{2.303RT} + logA ------(6)$$

Where T is the absolute temperature, A is the Arrhenius constant, R is the gas constant (8.314 J/mol/K), and C_R is the corrosion rate.

Using equation (7) below, the value of the heat of adsorption Q_{ads} was also determined.

$$Q_{ads} = 2.303R\left[\left(\log\frac{\theta_2}{1-\theta_2}\right) - \log\left(\frac{\theta_1}{1-\theta_1}\right)\right] \times \left(\frac{T_2T_1}{T_2-T_1}\right)\frac{J}{mol} --- (7) \text{ Onen } \textit{et al.} \text{ (2011)}$$

Where θ_1 and θ_2 are degrees of surface coverage at 303 K and 323 K.

Using the transition state equation (8), the standard enthalpy and standard entropy of activation were determined.

$$CR = \frac{RT}{Nh} exp^{\frac{\Delta S^*}{R}} exp^{\frac{-\Delta H^*}{RT}} - - - - - (8)$$

Where h is the plank's constant and N is the Avogadro's number, respectively. A plot of $log\frac{CR}{T}$ $verse\frac{1}{T}$ gave a straight lines with a slope of$\frac{-\Delta H^*}{2.303R}$ $and\ intercept\ of$ [log$\left(\frac{R}{Nh}\right)$ + $\frac{\Delta S^*}{2.303R}$], from which the activation thermodynamics parameters (ΔH^*) $and(\Delta S^*)$were calculated.

Adsorption Isotherm

In order to comprehend the mechanism underlying the prevention of corrosion reactions, adsorption isotherms are crucial. The plant extract's adsorption behavior as a green corrosion inhibitor was examined using the Flory-Huggins, Frumkin, Temkin and Langmuir adsorption isotherms (Joseph and Vincent, 2012).

Langmuir adsorption isotherms

Langmuir adsorption isotherm is expressed according to Equation (9)

$$\frac{C}{\theta} = \frac{1}{K} + C \qquad (9)$$

Where C is the concentration of inhibitor, K is the adsorption equilibrium constant and θ is the surface coverage of the inhibitor. Taking logarithm of both side of equation (9)

$$log\frac{C}{\theta} = logC - logK \qquad (10)$$

Temkin adsorption isotherms

For the Temkin isotherm, the degree of surface coverage (θ) is related to an inhibitor concentration (C) is given in the equation (11) below;

$$\exp(-2a\theta) = KC \qquad (11)$$

Where K is the adsorption equilibrium constant and a, is the attractive parameter. Rearranging and taking logarithm of both side of equation (11)

$$\theta = \frac{-2.303logK}{2a} - \frac{2.303logC}{2a} \qquad (12)$$

Frumkin adsorption isotherms

The Frumkin adsorption isotherm is given by equation 13

$$log\left[[C] \times \left(\frac{\theta}{1-\theta}\right)\right] = 2.303logK + 2\alpha\theta \qquad (13)$$

Where K is the adsorption-desorption constant and α is the lateral interaction term describing the interaction in adsorbed layer.

Flory-Huggins adsorption isotherms

The Flory-Huggins adsorption isotherm is given by equation 14 (Eddy *et al.,* 2009).

$$\log\left(\frac{\theta}{C}\right) = logK + xlog(1-\theta) \qquad (14)$$

where x is the size parameter and is a measure of the number of adsorbed water molecules substituted by a given inhibitor molecule.

Fourier Transformation Infrared (FTIR)

The leave extract of Christ's thorn was characterized by Fourier-transform infrared (FT-IR) spectroscopy. A Perkin Elmer Spectrum Happ-Genzel FT-IR was used to obtain infrared emission spectra with wave number ranging from 500 to 4000 cm^{-1} to identify the functional group present in the ethanolic extract (Dipak*et al.,* 2020).

Scanning Electron Microscope (SEM)

The mild steel coupon was immersed for 2 hours in 0.9 M HCl solution containing optimum concentrations (320 pmm Christ thorn), after 2 hours, the coupons were taken out and dried. The surface morphology of the mild steel was examined at 15kV and 500x using the JEOL (JSM 6390) Scanning electronic microscope (Yadev *et al.,* 2015).

RESULTS AND DISCUSSION

Phytochemical Analysis

The identification of the active phytochemical in the crude extracts is shown in Table 1 below. The findings showed that all the phytochemicals on the list that were screened for were present in leave extract, with the exception of anthraquinones. Generally speaking, the leaves of Christ Thorn (CT) plants contain some concentration of phytochemicals, including metabolites such as organic acids, phenolic compounds, saponins, alkaloids, flavonoids, terpenoids, and carbohydrates (Mohammad *et al.,* 2020). Christ Thorn leaves can be used to identify active substances through a bio-guided phytochemical investigation. These complex structures physical, chemical, and biological characteristics prevent metal and alloy corrosion (Anindita and Bikramjit, 2017).

Table: 1 Phytochemical Screening of Christ Thorn Leaves

Phytochemicals	Qualitative Analysis
Alkaloids	+
Anthraquinones	-
Cardia glycosides	+
Carbohydrates	+
Flavonoids	+
Saponins	+
Steroids/Triterpenes	+
Tannins	+
Terpenoids	+

Key + indicates the presence of phytochemicals, - indicates the presence of phytochemicals

Weight Loss Analysis

The figures below show the results obtained from weight loss analysis, the results indicated that increase in weight loss with increase in time of immersion.

Effect of Inhibitor Concentration

The effect of inhibitor of Christ Thorn leave extract was studied at different concentrations (0.20 g/L, 0.40 g/L, 0.60 g/L and 0.80 g/L), at 0.90 M HCl concentrations at 303 and 323 K temperatures for the corrosion inhibition of mild steel metal. The result from Figures 1 and 2 below indicates that the weight loss of mild steel metal decreases with an increase in inhibitor concentrations and increases with an increase in contact time. The results in Table 2 below showed that the corrosion rate C_R decreases with an increase in the concentration of inhibitors. Moreover, surface coverage (θ) and percentage inhibitor efficiency (% IE) increase with increasing concentrations of the inhibitor (Abdallah *et al.,* 2014).

Thermodynamic/Adsorption Parameters

Activation Energy and Heat of Adsorption

The activation energies were calculated using the Arrhenius equation and the heat of adsorption was also using equation 7 and the results are presented in Table 3. The results indicate that addition of leave extracts up to 0.80 g/L in 0.90 M HCl solutions increases the activation energy from 37.433 to 55.862 kJmol^{-1}, while the heat of adsorption from -22.04 to -17.73 kJmol^{-1} for the leaves extract while the calculated Ea for the blank solution is 34.178 kJmol^{-1}, this is because the amounts of the extracts are more which hindered the reaction process and the values of the plant extract solutions found to be greater than those that found in blank, which suggests that the formation of adsorption film of physical electrostatic reaction. The values of heat of adsorption in table 3 below indicate that the adsorption of ethanolic extracts of Christ Thorn leaves on the mild steel surface is exothermic (Abdallah *et al.*, 2014; Al-Otaibi *et al.*, 2014).

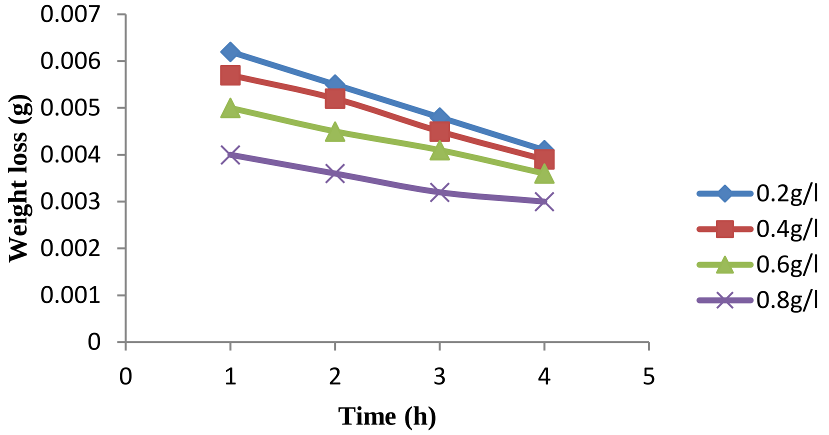

Fig. 1: Plot for weight loss of Christ Thorn Leave at 0.9M HCl at 303 K

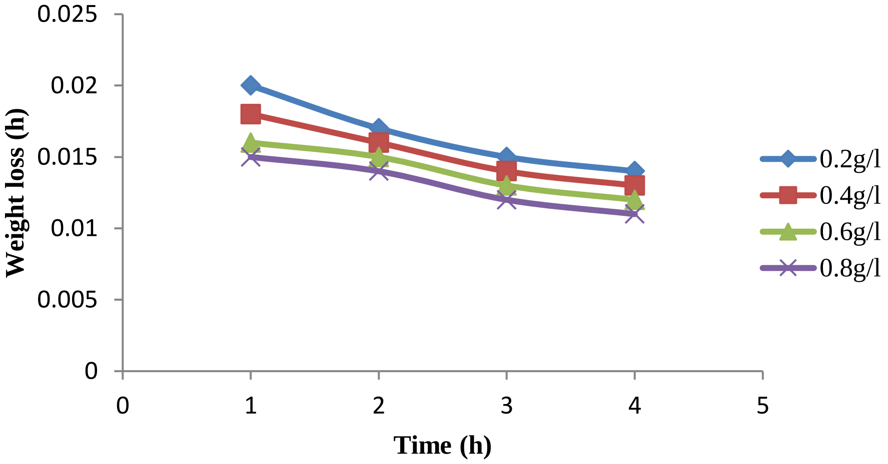

Fig. 2: Plot for weight loss of Christ Thorn Leave at 0.9M HCl at 323 K

Table 2: Surface Coverage θ, Percentage Inhibition Efficiency (%IE) of the Leave Extract and Corrosion Rate C_R Obtained from Weight Loss Experiment

Temperature	Inhibitor	Θ	%IE	C_R
303 K	Blank	-	-	0.385
	0.2 g/L	0.4715	47.15	0.342
	0.4 g/L	0.5106	51.06	0.276
	0.6 g/L	0.5834	58.34	0.204
	0.8 g/L	0.6004	60.04	0.169
323 K	Blank	-	-	0.876
	0.2 g/L	0.4431	44.31	0.774
	0.4 g/L	0.4763	47.63	0.613
	0.6 g/L	0.5168	51.68	0.567
	0.8 g/L	0.5409	54.09	0.518

Enthalpy ΔHα and Entropy ΔSα of Activation

Activation enthalpy ΔHα and entropy ΔSα of the corrosion process were calculated at various inhibitor concentrations and the results Christ Thorn were presented in Table 4. The values of activation enthalpy in 0.00 g/L and 0.80 g/L inhibitors (uninhibited and leaves extract) in 0.90 M HCl solution are 13.676, 15.098 – 20.315 kJmol^{-1} respectively, which are all positive in the presence and absence of an inhibitor and the inhibited values are greater than uninhibited values, these reflect that the dissolution process is endothermic (Abdallah *et al.,* 2014) while the values of an uninhibited entropies of adsorption in 0.0 g/L is 1.486, for the leave extract values from 0.20 – 0.80 g/L are 1.422 - 1.042 kJmol^{-1}, which indicates that activated complex in the rate-determining step represents association rather than dissociation, which means that there is reduction is disorderliness on going from reactant to activated complex (Abdallah *et al.,* 2014; Ghalib *et al.,* 2020). Generally, an exothermic adsorption process suggests either physisorption or chemisorption while an endothermic process is attributed to chemisorption only. Generally, enthalpy values up to 70 kJmol^{-1} are related to the electrostatic interactions between charged molecules and charged metal (physisorption) while those around 100 kJmol^{-1} or higher are attributed to chemisorption. In this case of Christ's thorn leave extract, the absolute values of enthalpy are relatively low, approaching those typical of physisorption and the values of ΔGα in the presence of christ thorn extracts are large and negative and are accompanied by exothermic adsorption process (Abdallah *et al.,* 2014; Ghalib *et al.,* 2020; Umoren *et al.,* 2020).

Free Energy of Adsorption ΔG_{ads}

The free energy of adsorption was calculated and the values of ΔGads for the tested isotherm in Table 5 revealed that for the leave extract, -16.034 is the lowest and -11.208 is the highest in magnitude which are negatively less than -40 kJmol^{-1} as required for the mechanism of chemical adsorption take place. Consequently, the adsorption process of Christ's thorn leave extract on the MS surface is spontaneous and the mechanism of physical adsorption is considered to be the nature of metal-inhibitor interaction. Any ΔGads which is negatively less than the threshold value -40 kJmol^{-1} required for the mechanism of chemical adsorption to take place, the adsorption process is physisorption (Abdallah *et al.,* 2014).

Adsorption Isotherms

The four selected models of adsorption isotherms: Langmuir, Temkin, Frumikn and Flory-Huggins isotherms were used for this study to describe the model of adsorption and to establish the best adsorption isotherm(s). The results of this investigation are presented in Table 5. Their regression coefficient (R^2) values were used to determine the best-fit isotherm among them. The best fit was considered by taking the values of R^2 which will give exactly or nearly equal unity at that temperature. From the table 5 below, the application of Langmuir isotherm to the adsorption of extracts of Christ Thorn leave on the surface of MS indicated that there is no interaction between the adsorbate and adsorbent (Ghalib *et al.,* 2020; Joseph *et al.,* 2012). The adsorption parameters obtained from the Temkin adsorption isotherm are recorded and the values of the attractive parameter are all negative in all cases which indicate that repulsion exists in the adsorption layer. The Frumkin adsorption isotherms parameter exhibits a negative value, indicating that the inhibitor exhibits an attractive behavior on the mild steel surface. The values of the size parameter of Flory-Huggins are all positive which indicates that the absorbed species of ethanolic extracts are compact since they could displace more than one water molecule from the mild steel surface (Abdallah *et al.,* 2014).

Table 3: Activation Energy Ea (kJmol^{-1}) and Heat of Adsorption Q_{Ads} (kJmol^{-1}) at Various Inhibitors of Leave Extracts

Inhibitor Conc	Activation Energy Ea	Heat of Adsorption Q_{Ads}
0.00	34.178	-
0.20	37.433	-22.04
0.40	44.259	-20.01
0.60	49.438	-18.53
0.80	55.862	-17.73

Surface Morphology Studies

Fourier Transform Infrared Spectroscopic Analysis (FT-IR)

The FT-IR spectra of the Christ's thorn leave extracts and corroded MS sample (corrosion product), inhibited sample were carried out and results as presented in Figure 3 to 5.

Table 4: Activation Enthalpy ΔH (kJmol^{-1}) and Entropy ΔS (kJmol^{-1}) at Various Inhibitors of Leave Extracts

Inhibitor Concentration gL^{-1}	Thermodynamic Parameter	
	ΔH	ΔS
0.00	13.676	1.486
0.20	15.098	1.422
0.40	18.062	1.192
0.60	20.092	1.076
0.80	20.315	1.042

Table 5: Adsorption parameters for adsorption of ethanolic extract of Christ Thorn leave on mild steel surface at 303 and 323 K

Isotherms	Temperatures	K_{ads}	ΔG (kJmol^{-1}	R^2
Langmuir	303 K	7.9086	-16.034	0.9971
	323K	6.7889	-15.330	0.9996
Temkin	303 K	-12.302	-17.011	0.7436
	323K	-10.147	-16.433	0.9028
Frumkin	303 K	-3.6446	-13.805	0.7982
	323K	-3.0755	-13.378	0.9213
Flory-Huggins	303 K	2.4927	-11.565	0.7882
	323 K	1.9356	-11.208	0.8071

In Figure 3, the FT-IR spectrum of Christ's thorn leaves reveal a broad band at 3279 cm^{-1}, indicating -OH or N-H stretching vibration. Other notable peaks include those at 2921-2854 cm^{-1} for -CH aliphatic compounds, 1704 cm^{-1} for -C=O carbonyl, and 1607-1514 cm^{-1} for aromatic -C=O stretching. Various other bands correspond to different functional groups. The FTIR spectrum of the corrosion product (Figure 4) formed on mild steel immersed in an acid solution of Christ's thorn leaves shows changes, such as a downshift from 3279 to 3231 cm^{-1} and an increase in the C=O stretching frequency from 1607 to 1640 cm^{-1}. Comparing with Figure 6, these shifts suggest alterations in hydroxyl, amino, and carbonyl groups. These shifts in frequencies indicate physical interactions between the metal surface and inhibitor molecules. Some functional groups disappear during the reactions, reinforcing the notion of a protective interaction between the extracts and the mild steel surface (Salwa *et al.*, 2022).

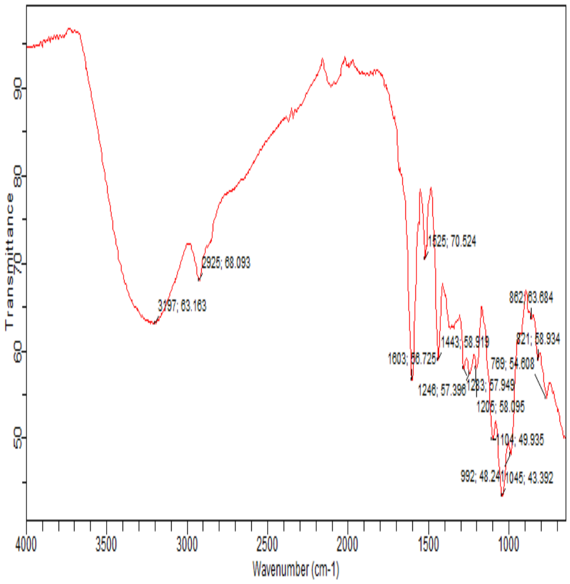

Fig. 3: FT-IR Spectrum of Christ

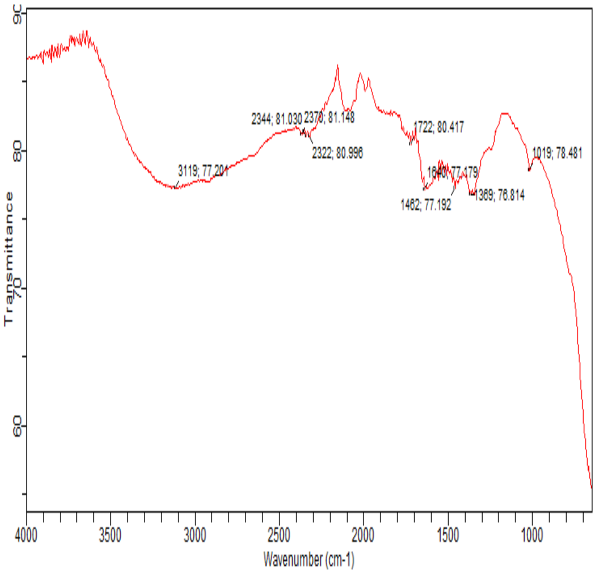

Fig. 4: FT-IR Spectrum of Inhibited thorn Leaves Extract Product

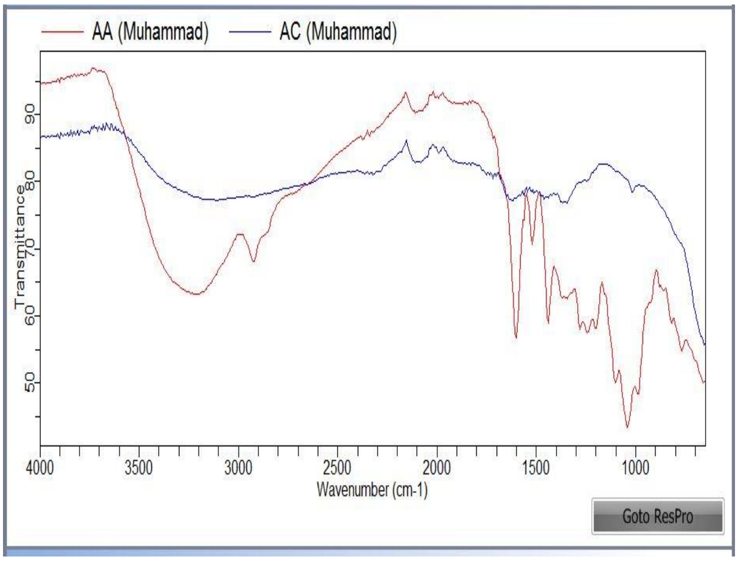

Fig. 5: Overlap spectrum of Christ thorn Leave and Inhibited Product

Scanning Electron Microscope (SEM)

Investigations for the surfaces morphology of fresh MS specimen, corroded MS specimen and inhibited MS specimen in 0.9 M HCl at 323 K were carried out by scanning electron microscopy (SEM) after immersion for 4 hrs, the SEM images were presented in the Figures below. The Figure 7 revealed severe corrosion on the metal surfaces in the uninhibited solution, indicating uniform damage characterized by cracks across the entire metal surface. Figures 8 to 11 demonstrate that the metal surfaces were effectively inhibited by the plant extracts of Christ Thorn leaves, with Figure 11 displaying the most protected surface due to the higher concentration of inhibitors (0.80 g/L) employed. This evidence supports the conclusion that the application of Christ Thorn leave extracts to acid solutions shields MS metal from corrosion by forming a protective layer on the surface through the adsorption of the extracts onto the MS surface. Hence, Christ Thorn leaves extract can be considered effective in controlling the corrosion of MS in acidic environments (Karthik *et al.*, 2015).

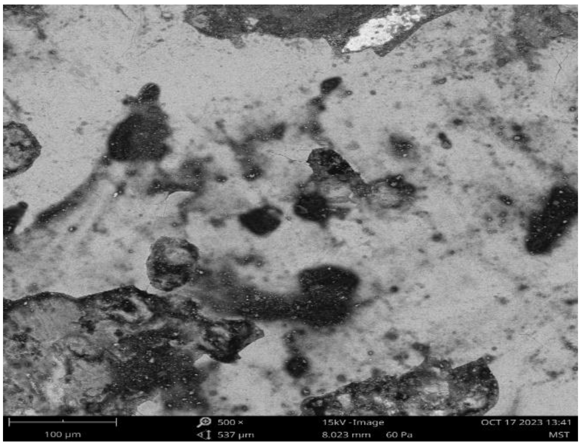

Fig 6: Surfaces Morphology of Fresh MS

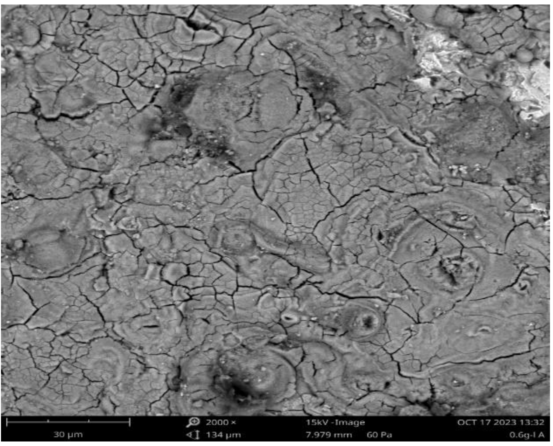

Fig 7: Corroded MS Specimen in 0.9 M Specimen HCl at 323 K

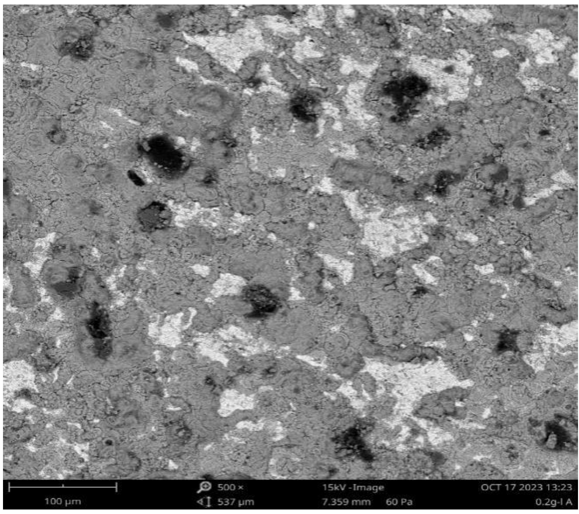

Fig. 8: Inhibited MS Specimen on Christ Thorn Leave in 0.20 g/L at 323 K

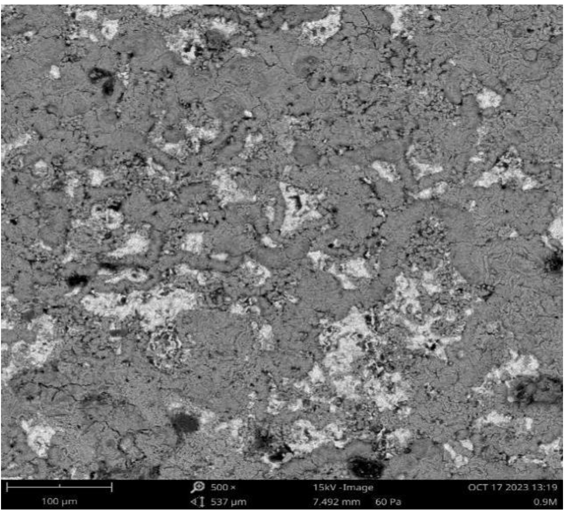

Fig. 9: Inhibited MS Specimen on Christ Thorn leave in 0.40 g/L at 323 K

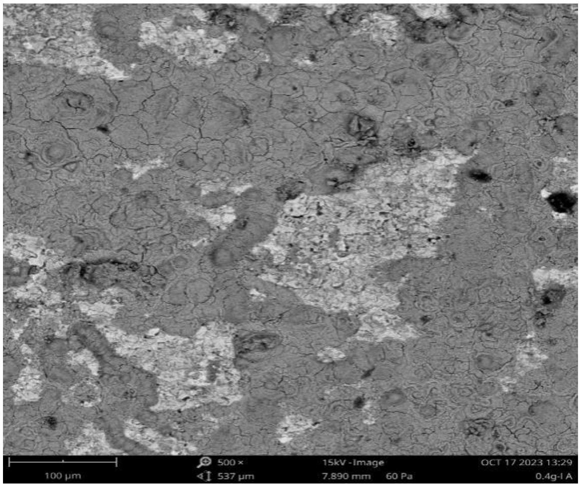

Fig. 10: Inhibited MS Specimen on Christ in 0.60 g/L at 323 K

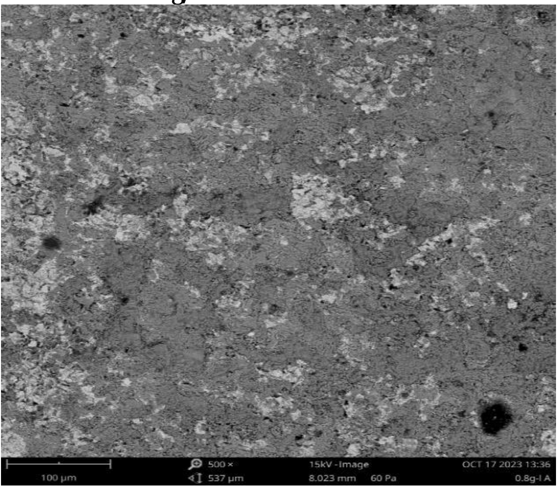

Fig. 11: Inhibited MS Specimen on Christ Thorn Thorn leave in 0.80 g/L at 323 K

CONCLUSION

The investigation involved the examination of extracts from leaves of Christ's thorn, revealing the presence of active phytochemicals. These compounds were identified as effective inhibitors, demonstrating the ability to impede the corrosion of mild steel in hydrochloric acid (HCl) solutions. The study observed a correlation between the concentration of plant extracts and inhibiting efficiency (%IE), with an increase in concentration leading to higher %IE. However, %IE exhibited a decline with rising temperatures, prolonged immersion times. Scanning electron microscopy (SEM) images illustrated the protective effect of the plant extracts on the metal surface under acidic conditions. Further analysis, including Fourier-transform infrared spectroscopy (FT-IR), kinetics, and thermodynamic parameters, elucidated the physical adsorption mechanism. Notably, the Langmuir adsorption isotherm model provided the best fit, indicating the nature of adsorption.

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