

Corrosion inhibition of carbon steel in acidic medium by Grapefruit oil extract

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Abstract

Carbon steel is widely used in various fields such as the field of agri-food their use in tins and materials used in the food producing seen this material and susceptible to corrosion phenomena are in their environmental which produces toxic compounds in the environmental medium of this material. To this end we investigated in this direction by study of the inhibition of the phenomenon of corrosion of this material. Grapefruit oil extract (GO) was tested as corrosion inhibitor for mild steel in 1M HCl. Electrochemical impedance spectroscopy (EIS) method are used. The inhibiting action increases with the concentration of the tested inhibitor. The highest efficiency 86.15% is obtained at 1g/L of GO. Effect of temperature is also studied between 298 K and 328 K; GO presents best inhibition efficiency even at elevated temperature, GO is adsorbed on the steel surface according to a Langmuir isotherm adsorption model. The thermodynamic data of activation are determined. In this study fatty acid composition contents of Grapefruit were determined. The used methods indicate that the Grapefruit oil extract is a good inhibitor in 1.0 M HCl. Some thermodynamic functions of the carbon steel dissolution process in acid medium were also determined and discussed.

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1. Introduction

Alloys steel is the well-known materials used in various industrial applications and highly susceptible to dissolution problem in aqueous solutions and especially in acidic mediums [1-4]. Inhibitors are used in these processes to control the metal dissolution. Hydrochloric acid is widely used in the pickling of steel and different steel based alloys [5-6]. One way of protecting steel from corrosion is to use corrosion inhibitors. Organic compounds containing heteroatoms are commonly used to reduce the corrosion attack on steel in acidic media [7-20]. The recent trend is towards environmentally friendly inhibitors. Most of the natural products are non-toxic, biodegradable and readily available in plenty. These advantages have incited us to draw a large part of program of our laboratory to examine natural substances as corrosion inhibitors such as: *Juniper oxycedrus extract* [21], *Avogado Nuts Extract* [22], *Chenopodium Ambrorsioides Extracts* [23-24], *Anemone coronaria Extract* [25], *Green Tea Extract* [26], *Oil of Citrus Leaves* [27], *Rosemary oil* [28-30], *Harmal Extract* [31], *Nigella Sativa Oil* [32], *Ylang Ylang Oil* [33], *Natural linseed Oil* [34], *anise oil* [35], *Artemisia* [36-38] and *Kaempferol* [39]. Grapefruit are among the most import tree fruit crops in the Mediterranean countries and their production and consumption have increased considerably in recent years. The present work was established to study the corrosion inhibition of C38 steel in 1M HCl solution by Grapefruit oil extract (GO) as corrosion inhibitor using electrochemical impedance spectroscopy (EIS).

2-Materials and methods.

2-1.Electrochemical tests

The electrochemical study was carried out using a potentiostat PGZ100 piloted by Voltamaster software. This potentiostat is connected to a cell with three electrode thermostats with double wall (Tacussel Standard CEC/TH). A saturated calomel electrode (SCE) and platinum electrode were used as reference and auxiliary electrodes, respectively. Potentiodynamic polarization curves were plotted at a polarization scan rate of 1 mV/s. Before all experiments, the potential was stabilized at free potential during 30 min. The polarization curves are obtained from -800 mV to -200 mV/SCE at 298 K. In order to investigate the effects of temperature and immersion time on the inhibitor performance, some test were carried out in a temperature range 298-328 K. The electrochemical impedance spectroscopy (EIS) measurements are carried out with the electrochemical system (Tacussel), which included a digital potentiostat model Voltalab PGZ100 computer at E_{corr} after immersion in solution. After the determination of steady-state current at a corrosion potential, sine wave voltage (10 mV) peak to peak, at frequencies between 100 kHz and 10 mHz are superimposed on the rest potential. Computer programs automatically controlled the measurements performed at rest potentials after 0.5 hour of exposure at 298 K. Nyquist plots were made from these experiments. The best semicircle can be fit through the data points in the Nyquist plot using a non-linear least square fit so as to give the intersections with the x-axis. The charge transfer resistance (R_{ct}) was determined from Nyquist plots and double layer capacitance (C_{dl}) was calculated from CPE parameters of the equivalent circuit deduced using Zview software. In this case the percentage protection efficiency $\eta_{\text{EIS}} (\%)$ is can be calculated by the value of the charge transfer resistance (R_{ct}). Where R_{ct}^0 and R_{ct} were the polarization resistance of uninhibited and inhibited solutions, respectively.

$$\eta_{\text{EIS}} \% = \frac{R_{\text{ct}} - R_{\text{ct}}^0}{R_{\text{ct}}} \times 100 \quad (1)$$

3-Results and discussion.

3-1. Electrochemical impedance spectroscopy measurements

The effect of addition of the **GO** as inhibitor on the corrosion behavior of carbon steel in 1.0 M HCl solution was studied by electrochemical impedance spectroscopy measurements at 298 K after at 30 min of immersion at open circuit potential (Fig. 1). The values of the electrochemical parameters that can be extracted from the Nyquist diagrams are collected in Table 1, Nyquist plots obtained for different concentrations of **GO** are similar with the shape of blank solution; they exhibit single depressed semicircles across the frequency range studied, which denotes that the corrosion process was controlled by charge transfer resistance [39]. De size of semicircles increases with concentration of **GO**. This depression can be attributed to the roughness of electrode surface and other inhomogeneity of the solid alloy caused by their dissolution during the attainment of corrosion potential [40]. The Nyquist plots were analysed with Zview 2 software by fitting the experimental data to a simple equivalent circuit model (Fig. 3) [41]. The fitted parameters including; the solution resistance R_s , the charge transfer resistance (R_{ct}) and the constant phase element (CPE) are tabulated in Table 1. The value of double layer capacitance (C_{dl}) may be determined using the following equation (2). Where n is a CPE exponent which can be used as a gauge for the heterogeneity or roughness of the surface and Q is the CPE constant (in $\Omega^{-1} S^n cm^{-2}$), R_{ct} is the charge transfer resistance.

$$C_{dl} = \sqrt[n]{Q \cdot R_{ct}^{1-n}} \quad (2)$$

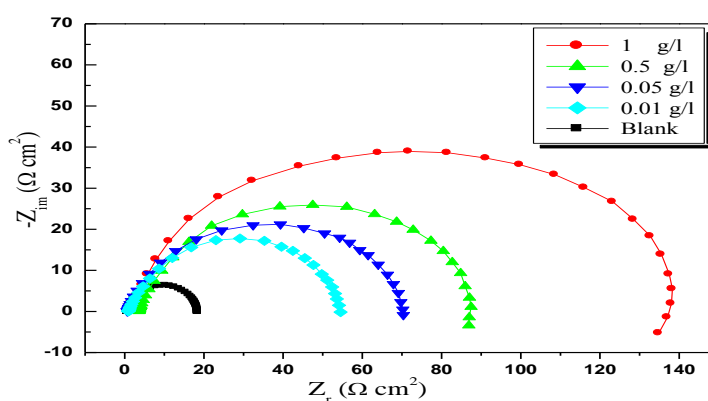


Figure 1. Nyquist diagrams for carbon steel in 1.0 M HCl containing different concentrations of Grapefruit oil extract at 298 K.

Table 1. Impedance parameters for corrosion of carbon steel in 1.0 M HCl in the absence and presence of different concentrations of Grapefruit oil extract at 298 K.

	Concentration (g/L)	R_{ct} ($\Omega \times cm^2$)	n	$Q \times 10^{-4}$ ($s^n / \Omega \times cm^2$)	C_{dl} ($\mu F / cm^2$)	η_{EIS} (%)	Θ
Blank	-	18	0.79	2.092	47.75	-	-
GO	1.00	130	0.76	0.44	8.67	86.15	0.86
	0.50	85	0.73	0.68	10.98	78.82	0.78
	0.05	68	0.74	0.75	11.77	73.52	0.73
	0.01	55	0.73	1.07	16.12	67.72	0.67

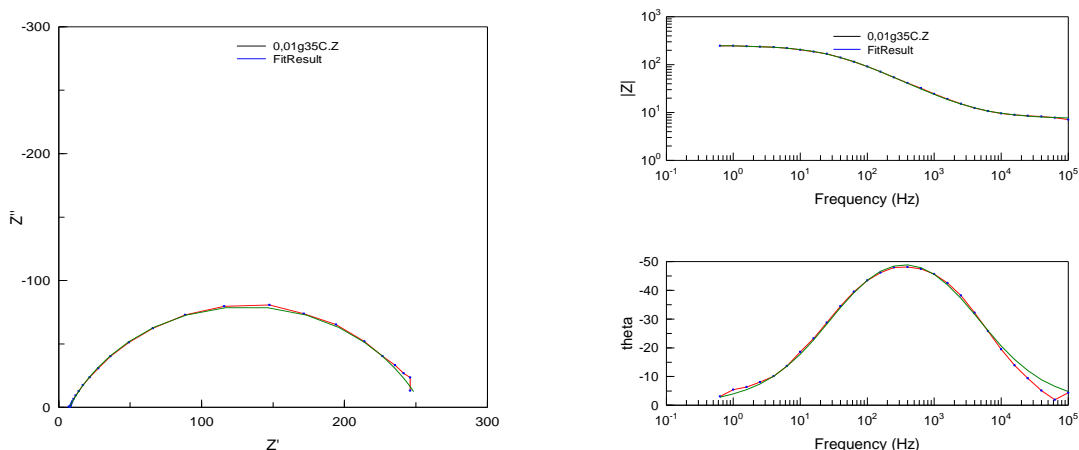


Figure 2: EIS Nyquist (a) and Bode (b) plots for carbon steel / 1M HCl + (**GO**) interface

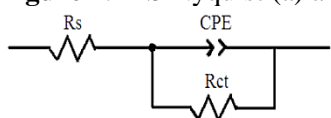


Figure 3. Equivalent electrical circuit corresponding to the corrosion process on the carbon steel in hydrochloric acid.

As can be seen from table.1 the increase in resistance in the presence of **GO**, compared to HCl, alone is related to the corrosion inhibition effect of the molecules. The value of C_{dl} decreases in the presence of this inhibitor. The decrease in C_{dl} , which can result from a decrease in local dielectric constant and/or an increase in the thickness of the electric double layer [42], suggested that **GO** molecules function by adsorption at the metal/solution interface. The resistance of charge transfer, R_{ct} , increase gradually with concentration, which is accompanied of decrease in values of the double layer capacitance C_{dl} , suggests that the adsorption surface increases with concentration of the inhibitor and there by supply excellent barrier towards charge transfer reactions at the metal–solution interface [43].

3-2. Effect of temperature

The study of the temperature effect on the inhibition behavior is very important for understanding the adsorption of inhibitor molecules. Figs. 4 and 5 present the impedance spectroscopy (EIS) measurements for carbon steel in the absence and presence of different concentration of the studied inhibitor in 1.0 M HCl at different temperatures. The electrochemical parameters deduced from this investigate are listed in Table 2.

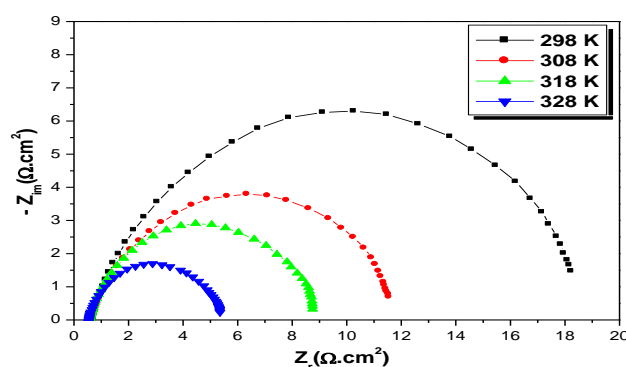


Figure 4 . Nyquist diagrams for carbon steel in 1.0 M HCl at different temperatures.

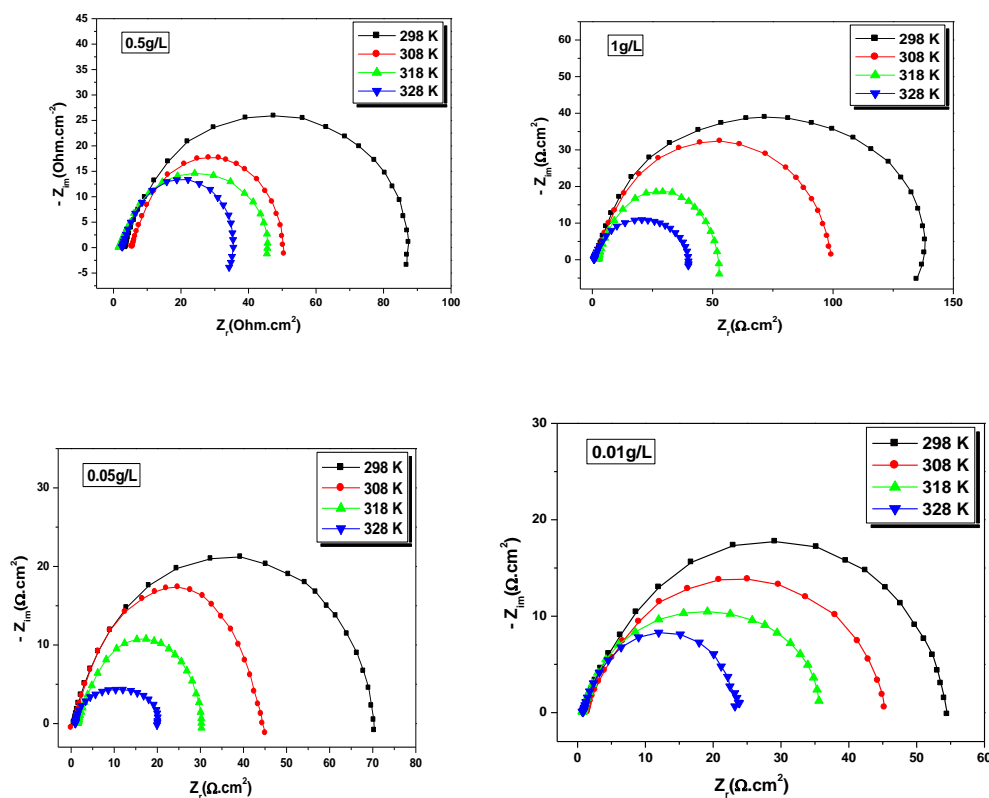


Figure 5. Nyquist diagrams for carbon steel in 1 M HCl + 1g/L;0.5 g/L;0.05 g/L;0.01 g/L of GO at different Temperatures.

Also inhibition efficiency of GO increased with increase in temperature. Values of R_{ct} were employed to calculate values of the corrosion current density (I_{corr}) at various temperatures in absence and presence of GO using the following equation [44]:

$$I_{corr} = R.T.(Z.F.R_{ct})^{-1} \quad (3)$$

Where R is the universal gas constant ($R = 8.31 \text{ J K}^{-1}\text{mol}^{-1}$), T is the absolute temperature, z is the valence of iron ($z = 2$), F is the Faraday constant ($F = 96.485 \text{ Coulomb}$) and R_{ct} is the charge transfer resistance.

Fig. 6 also shows that the corrosion reaction can be regarded as an Arrhenius-type process (Eq 5). The activation parameters for the studied system (E_a , ΔH_a and ΔS_a) were estimated from the Arrhenius equation and transition state equation (Eq 4-5):

$$I_{corr} = A \exp\left(\frac{-E_a}{RT}\right) \quad (4)$$

$$I_{corr} = \frac{RT}{Nh} \exp\left(\frac{-\Delta S_a}{RT}\right) \exp\left(\frac{-\Delta H_a}{RT}\right) \quad (5)$$

Where A is Arrhenius factor, E_a is the apparent activation corrosion energy, N is the Avogadro's number, h is the Plank's constant, and $-\Delta H_a$ and $-\Delta S_a$ are the enthalpy and the entropy changes of activation corrosion energies for the transition state complex. R is the perfect gas constant.

The apparent activation energy was determined from the slopes of $\ln I_{corr}$ vs $1/T$ graph depicted in Fig. 7.

Table 2. Thermodynamic parameters for the adsorption of GO in 1.0 M HCl on the carbon steel at different temperatures.

	Temp (K)	R_{ct} ($\Omega.cm^2$)	n	$Q \times 10^{-4}$ ($s^n/\Omega \times cm^2$)	C_{dl} ($\mu F/cm^2$)	E_{Ret} (%)
Blank	298	18	0.79	2.092	47.75	-
	308	11	0.77	2.69	7.30	-
	318	8	0.80	2.37	4.94	-
	328	5	0.81	2.17	3.77	-
1 g/L	298	130	0.76	0.44	8.67	86.15
	308	98	0.76	0.45	7.93	88.77
	318	52	0.76	0.38	5.35	84.61
	328	40	0.79	0.25	4.01	87.5
0.5g/L	298	85	0.81	0.68	21.1	78.82
	308	50	0.73	0.62	7.37	78
	318	45	0.80	0.34	6.72	82
	328	32	0.78	0.32	4.59	84
0.05g/L	298	68	0.74	0.75	11.77	73.52
	308	45	0.71	0.67	6.72	75.55
	318	30	0.77	0.38	5.44	73.33
	328	20	0.79	0.26	3.51	75
0.01g/L	298	55	0.73	1.07	16.12	67.72
	308	36	0.72	0.86	9.16	69.44
	318	24	0.78	0.53	8.08	66.6
	328	16	0.78	0.52	3.32	68.75

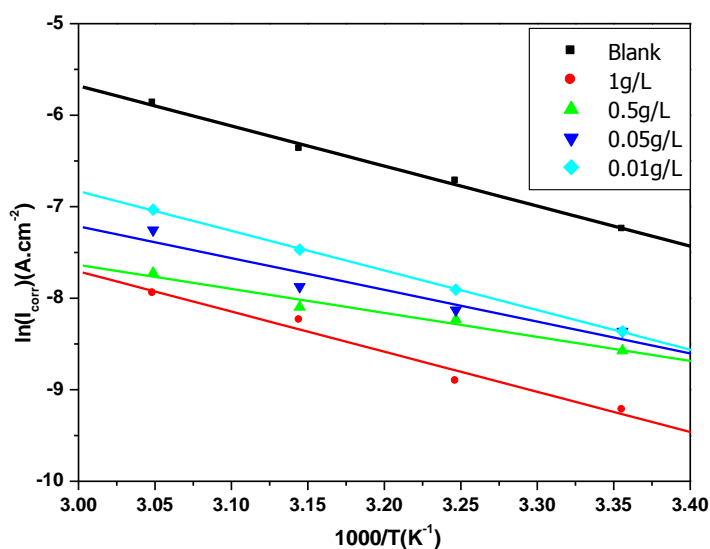


Figure 6. Arrhenius plots of C38 steel in 1M HCl with and without 1g/L, 0.5 g/L; 0.05 g/L; 0.01 g/L of GO

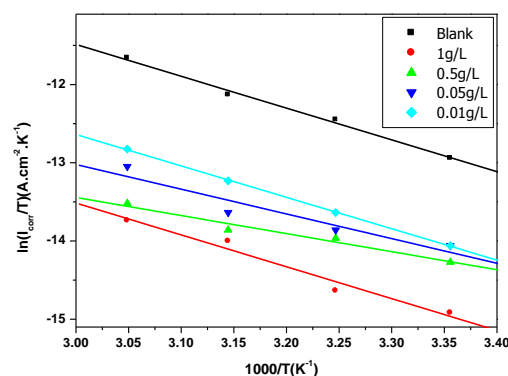


Figure 7 . Relation between $\ln(I_{corr}/T)$ and $1000/T$ at different temperatures.

A plot of $\ln(I_{corr}/T)$ against $1/T$ (Fig. 7) gave a straight line with slope $\Delta H_a/R$ and intercept $(\ln(R/N A h) + (\Delta S_a/R))$, from which the values of ΔH_a and ΔS_a were calculated and listed in Table 3.

Table 3. Activation parameters for the corrosion of C-steel in 1.0 M hydrochloric acid containing different concentrations of inhibitor GO.

	E_a (kJ/mol)	ΔH_a (kJ/mol)	ΔS_a (J/mol K)	$E_a - \Delta H_a^*$	ΔG_a (kJ/mol) (T=298K)
Blank	36.38	33.79	-191.53	2.60	93.45
1g/L	36.45	33.85	-208.19	2.60	98.49
0.5g/L	32.53	29.85	-218.57	2.60	97.66
0.05g/L	30.81	27.81	-221.51	2.60	96.81
0.01g/L	24.49	21.90	-241.20	2.60	96.36

From this table 3: The positive sign of the enthalpy ΔH_a reflects the endothermic nature of the steel dissolution process, which means that the dissolution of steel is difficult [45]. In this act some authors [46-48] have attributed this result to the inhibitor species being physically adsorbed on the metal surface. We remark that E_a and ΔH_a values vary in the same way (Table 3). This result permit to verify the known thermodynamic reaction between the E_a and ΔH_a as shown in Table 3 [49]:

$$\Delta H_a = E_a - RT \quad (6)$$

The negative value of entropy ΔS_a in the presence of an inhibitor implies that the activated complex in the rate determining step represents an association, rather than a dissociation step, meaning that an increase in disordering takes place in going from reactants to the activated complex [50].

3-5. Adsorption isotherm

The adsorption behavior of the inhibitor molecules on electrode surface can be used to elucidate the inhibition mechanism of inhibitors. The surface coverage values (θ) determined by electrochemical impedance spectroscopy (EIS) (Table 1) at different concentrations of **GO** inhibitor in acidic solution were used to explain the excellent

adsorption isotherm, consequently, establish the adsorption process from the experimental results obtained. To investigate the inhibitor adsorption mechanism, several adsorption isotherms were fitted and the Langmuir adsorption isotherm gives the best description of the adsorption behavior of the inhibitor on carbon steel electrode according to Langmuir adsorption isotherm equation:

$$\frac{C_{inh}}{\theta} = \frac{1}{K_{ads}} + C_{inh} \quad (7)$$

Where C_{inh} is the inhibitor concentration and K_{ads} is the adsorptive equilibrium constant. The linear relationship of C_{inh}/θ versus C_{inh} in different concentrations of inhibitor presented in Fig.8, possesses a slope near to unit with the correlation coefficient R^2 that is close to 1, thus, the adsorption of **GO** on steel surface obeys the Langmuir adsorption isotherm. The corrosion inhibition can be attributed to the adsorption of the Grapefruit oil extract at the steel/acid solution interface. The highest inhibition efficiency 86.15 % was obtained at 1g/L of Grapefruit oil extract.

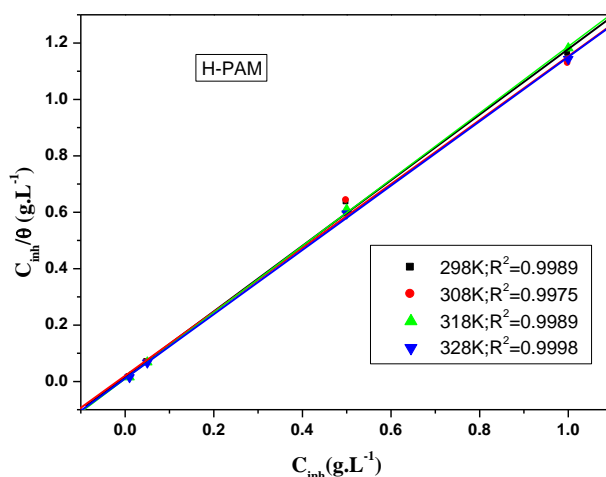


Figure 8. Langmuir adsorption Grapefruit oil extract on the carbon steel surface in 1.0 M HCl solution at different Temperatures.

4. Conclusion.

This study demonstrates that Grapefruit oil extract (**GO**) is a good inhibitor of corrosion for carbon steel in corrosive environment. The adsorption of the tested inhibitor on interface between the carbon steel and molar hydrochloric acid obeys the Langmuir adsorption isotherm model. The inhibitory effect of **GO** on the electrode surface increases with concentration of inhibitor and decrease slightly with temperature. EIS measurement shows a single depressed semicircle observed from high to low frequencies region for uninhibited and inhibited solution illustrate that the corrosion process was controlled by charge transfer resistance. The increasing of charge transfer resistance (R_{ct}) and decreasing of double layer capacitance (C_{dl}) in the presence of inhibitor can be suggested by the adsorption the inhibitor molecules on the surface of carbon steel.

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