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Inhibition of steel corrosion in hydrochloric acid solution by chamomile extract

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ABSTRACT

Chamomile extract (CE) was tested as corrosion inhibitor for C38 steel in 1 M HCl using potentiodynamic polarization, electrochemical impedance spectroscopy (EIS) and scanning electron microscope (SEM) studies. The effect of temperature on the corrosion behavior of C38 steel in 1 M HCl with addition of plant extracts was studied in the temperature range of 298–328 K. Inhibition efficiency of 88% was achieved with 7g/L CE at 298 K. It is evident from the results of this study that CE inhibits the corrosion in 1 M HCl through adsorption process following Langmuir adsorption isotherm. The protection efficiency increased with increase in inhibitor concentration but decreases with temperatures. The Nyquist plots showed that on increasing CE concentration, increases charge transfer resistance and decreases double layer capacitance. The mixed mode of action exhibited by the inhibitor was confirmed by the polarization studies while SEM analysis substantiated the formation of protective layer over the C38steel surface. The values of inhibition efficiency obtained from different techniques measurements are in good agreement.

Keywords: Polarisation, EIS, SEM, Corrosion inhibition, Steel, chamomile extract, acid medium

INTRODUCTION

Acid solutions are used in industry to remove mill scale from metallic surfaces [1–3]. Hydrochloric acid and sulfuric acid are widely used for this purpose. However, most acid media cause metal corrosion. To ovoid the attack of acid to the bulk metal, inhibitors are generally added. Organic compounds containing heteroatoms are commonly used to reduce the corrosion attack on steel in acidic media [4-14]. The use of non-toxic inhibitors called green or eco-friendly environmental inhibitors is one of the solutions possible to prevent the corrosion of the material. These advantages have incited us to draw a large part of program of our laboratory to examine natural substances as corrosion inhibitors such as: *prickly pear seed oil* [15], *Argan* oil [16, 17], *Argan* extract [18, 19], *Fennel* oil [20], *Rosemary oil* [21-23], *Thymus oil* [24, 25], *Lavender oil* [26], *Jojoba oil* [27], *Pennyroyal Mint oil* [28] and *Artemisia* [29-31]. All of which have been reported to be good inhibitors for metals and alloys in acidic solutions.

The objective of this study was to find an environmentally safe and inexpensive inhibitor (chamomile extract) of corrosion of C38 steel in acidic solution by polarization, electrochemical impedance spectroscopy (EIS) and scanning electron microscope (SEM) studies.

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. The surface area exposed to the electrolyte is 0.094 cm².

Potentiodynamic polarization curves were plotted at a polarization scan rate of 0.5 mV/s. Before all experiments, the potential was stabilized at free potential during 30 min. The polarisation curves are obtained from -800 mV to -400 mV at 298 K. The solution test is there after de-aerated by bubbling nitrogen. Gas bubbling is maintained prior and through the experiments. 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 without bubbling. 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. The impedance diagrams are given in the Nyquist representation. Experiments are repeated three times to ensure the reproducibility.

2.2 Solutions preparation

Sample of *Chamomile (Chamomilla recutita)* was collected in the food supermarket of Agadir, Morocco. Dried and pulping fruits were crushed. Stock solution of the chamomile extract was prepared by stirring cold weighed amounts of the chamomile plant for 24 h in 1 M HCl solution (The solution 1M HCl was prepared by dilution of analytical grade 37% HCl with double distilled water). The resulting solution was filtered. This extract was used to study the corrosion inhibition properties and to prepare the required concentrations. The solution tests are freshly prepared before each experiment. Experiments were carried out in triplicate to ensure the reproducibility.

2.3 Scanning electron microscopy (SEM)

Immersion corrosion analysis of carbon steel samples in the acidic solutions with and without the optimal concentration of the inhibitor was performed using SEM. Immediately after the corrosion tests, the samples were subjected to SEM studies to know the surface morphology. SEM Jeol JSM-5800 was used for the experiments.

RESULTS AND DISCUSSION

3.1 Electrochemical impedance spectroscopy measurements

The corrosion behaviour of steel in 1M HCl solution, in the absence and presence of CE, is also investigated by the EIS at 298 K after 30 min of immersion. The charge-transfer resistance (R_{ct}) values are calculated from the difference in impedance at lower and higher frequencies, as suggested by Tsuru et al [32]. The double layer capacitance (C_{dl}) and the frequency at which the imaginary component of the impedance is maximal (- Z_{max}) are found as represented in equation:

$$C_{dl} = \left(\frac{1}{\omega R_{l}}\right) \qquad \text{where } \omega = 2\pi f_{\text{max}} \tag{1}$$

The inhibition efficiency got from the charge transfer resistance is calculated by:

$$\% IE_{R_{ct}} = \left(1 - \frac{R_{ct}^{\circ}}{R_{ct}}\right)$$
(2)

where R_{ct} and R_{ct} are the charge transfer resistance in the absence and presence of different concentrations of inhibitor, respectively.

Nyquist representation of the EIS study of C38 steel in 1M HCl in absence and presence at differents concentrations of CE were presented in Fig. 1. The large capacitive loop attributed to the adsorption of the inhibitor molecule [33]. The existence of single semi circle showed the single charge transfer process during dissolution which is unaffected

by the presence of inhibitor molecules. Deviations of perfect circular shape are often refereed to the frequency dispersion of interfacial impedance which arises due to the roughness and other inhomogeneity of the surface [34, 35]. As seen from Table 1, the R_{ct} values of inhibited substrates are increased with the concentration of inhibitors. On the other hand, the values of C_{dl} are decreased with increase in inhibitor concentration which is most probably is due to the decrease in local dielectric constant and / or increase in thickness of the electrical double layer, suggests that CE act via adsorption at the metal / solution interface [36, 37].



Fig. 1. Nyquist plots of steel in 1 M HCl containing various concentrations of chamomile extract at Ecorr after 30 min of immersion.

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

Inhibitor	C (g/L)	Rt (k Ω .cm ²)	f _{max} (Hz)	$C_{dl}(\eta F/cm^2)$	$IE_{Rct}(\%)$
Blank	0.0	10	50	318	-
Chamomile Extract	3.0	24	40	165	66.7
	4.0	34	31	151	70.6
	5.0	46	25	138	78.2
	6.0	53	22	136	81.1
	7.0	84	14	135	88.0

3.2 Polarization measurements

3.2.1 Effect of concentration

Polarization measurements have been carried out in order to gain knowledge concerning the kinetics of the anodic and cathodic reactions. The anodic and cathodic current–potential curves are extrapolated up to their intersection at a point where corrosion current density (I_{corr}) and corrosion potential (E_{corr}) are obtained [38]. Table 2 shows the electrochemical parameters (I_{corr} , E_{corr} and b_c) obtained from Tafel plots for the C38 steel electrode in 1M HCl solution without and with different concentrations of MMI. The I_{corr} values were used to calculate the inhibition efficiency, E_{I} (%) (listed in Table 2), using the following equation :

$$E_{I}(\%) = \left(\frac{I_{corr} - I_{corr}}{I_{corr}}\right) \times 100 \tag{3}$$

Where I_{corr} and I_{corr} are uninhibited and inhibited corrosion current densities, respectively. Under the experimental conditions performed, the cathodic branch represents the hydrogen evolution reaction, while the anodic branch represents the iron dissolution reaction. They are determined by extrapolation of Tafel lines to the respective corrosion potentials. Some of the authors proposed the following mechanism for the corrosion of iron and steel in acid solution [39–41]:

$$Fe + A^{n-} \rightarrow (FeA^{n+})_{ads}$$

$$(FeA^{n-})_{ads} \rightarrow (FeA^{n-})_{ads} + ne^{-}$$
$$(FeA^{n-})_{ads} \rightarrow (FeA^{n+})_{ads} + ne^{-}$$
$$(FeA^{n+})_{ads} \rightarrow (Fe^{2+})_{ads} + A^{n-}$$

The cathodic hydrogen evolution $E_{a} + U^{+} \rightarrow (E_{a}U^{+})$

$$(FeH^{+})_{ads} \rightarrow (FeH^{-})_{ads}$$
$$(FeH^{+})_{ads} + ne^{-} \rightarrow (FeH)_{ads}$$
$$(FeH^{+})_{ads} + H^{+} + ne^{-} \rightarrow Fe + H_{2}$$

Inhibitor	C (g/L)	Ecorr(mV/SCE)	$I_{corr}(\mu A/cm^2)$	-b _c (mV/dec)	E _I (%)
Blank	0.0	-612	124	149	-
Chamomile Extract	3.0	-591	52	154	58.0
	4.0	-575	45	181	63.7
	5.0	-591	36	158	70.9
	6.0	-558	32	189	74.2
	7.0	-557	22	195	82.2

The inspection of results in Table 2 indicate that CE inhibits the corrosion process in the studied range of concentrations and E_I (%) increases with the concentration of the inhibitor, reaching its maximum value, 82.2%, at 7 g/L. The values of the cathodic Tafel lines, b_c , show slight changes with the addition of CE. This result means that the mechanism at the electrode reaction is not changed [42, 43].

Table 3. Polarisation parameters for C38 steel in 1 M HCl with various concentrations of CE at different temperatures.

T (K)	Conc	Ecorr (mV/SCE)	$I_{corr} (\mu A/cm^2)$	-b _c mV/dec)	E _I (%)	Θ
298	Blank	-612	124	149		
	3 g/L	-591	52	154	58.0	0.580
	4 g/L	-575	45	181	63.7	0.637
	5 g/L	-591	36	158	70.9	0.709
	6 g/L	-558	32	189	74.2	0.742
	7 g/L	-557	22	195	82.2	0.822
308	Blank	-544	177	132		
	3 g/L	-581	75	137	57.6	0.576
	4 g/L	-572	85	193	51.9	0.519
	5 g/L	-582	58	143	67.2	0.672
	6 g/L	-584	77	178	56.4	0.564
	7 g/L	-572	52	168	70.6	0.706
318	Blank	-545	280	133		
	3 g/L	-594	125	163	55.7	0.557
	4 g/L	-587	150	172	46.4	0.464
	5 g/L	-592	104	161	62.8	0.628
	6 g/L	-605	152	166	45.7	0.457
	7 g/L	-597	106	159	62.1	0.621
328	Blank	-537	435	140		
	3 g/L	-586	210	162	51.7	0.517
	4 g/L	-595	237	154	45.5	0.455
	5 g/L	-585	170	162	60.9	0.609
	6 g/L	-605	250	165	42.5	0.425
	7 g/L	-595	178	137	59.0	0.590

3.2.2 Effect of temperature

To evaluate the adsorption of CE and activation parameters of the corrosion processes of steel in acidic media, polarization measurements are investigated in the absence and presence of inhibitor and also in the range of temperature 298-328 K. Polarization curves for the C38 steel in 1 M HCl solution are shown in Figs. 2 and 3 in two different conditions, without different concentrations of CE and also in the presence of different concentrations of CE in the temperature range (298-328 K).

The numerical values of the variation of corrosion current density (I_{corr}), corrosion potential (E_{corr}), cathodic Tafel slope (b_c), and the degree of surface coverage (Θ) with the concentrations of inhibitor (CE) at all studied

temperatures are given in Table 3. These values were calculated from the intersection of the anodic and cathodic Tafel lines of the polarisation curve at E_{corr} . The surface coverage (Θ) was calculated using:

$$\Theta = E_I(\%) / 100 \tag{4}$$

The inhibition efficiency E_I (%) is given by equation 3

Close examination of Table 3 shows that an increase in temperature increases I_{corr} , while the addition of CE decreases the I_{corr} values across the temperature range. The results also indicate that the inhibition efficiencies increased with the concentration of inhibitor but decreased proportionally with temperature. Such behaviour can be interpreted on the basis that the inhibitor acts by adsorbing onto the metal surface, and an increase in temperature results in the desorption of some adsorbed inhibitor molecules, leading to a decrease in the inhibition efficiency [44].



Fig. 2. Potentiodynamic polarisation curves of steel in 1M HCl at different temperatures.



Fig. 3. Potentiodynamic polarisation curves of steel in 1M HCl in the presence of 7g/L of chamomile extract inhibitor at different temperatures.

Activation parameters such as the activation energy, E_a , the enthalpy of activation, ΔH_a^* , and the entropy of activation, ΔS_a^* , for both corrosion and corrosion inhibition of mild steel in 1 M HCl in the absence and presence of CE at different concentrations between 298 and 328 K were calculated from an Arrhenius-type plot (Eq. (4)) and the transition state (Eq. (5)) [45, 46]:

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

where I_{corr} is the corrosion current density (taken from averaged polarisation), A is the Arrhenius constant, E_a is the activation energy, and R is the universal gas constant.

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

where h is Planck's constant, N is Avagadro's number, ΔS_a^* is the entropy of activation and ΔH_a^* is the enthalpy of activation.

Plots of Ln (I_{corr}) vs. 1000/T and Ln (I_{corr} /T) vs. 1000/T gave straight lines with slopes of $-E_a/R$ and $-\Delta H_a^*/R$, respectively. Theintercepts were A and [Ln (R/Nh) + ($\Delta S_a^*/R$)] for the Arrhenius and transition state equations, respectively. Figs. 4 and 5 represent the data plots of Ln (I_{corr}) vs. 1000/T and Ln (I_{corr}/T) vs. 1000/T in the absence of CE at different concentrations. The calculated values of the activation energy E_a , the enthalpy of activation ΔH_a^* and the entropy of activation ΔS_a^* are tabulated in Table 4.



Fig. 4. Arrhenius plots of Ln(I_{corr}) versus 1000/T at various concentrations of *chamomile extract*.



Fig. 5. Variation of Ln (I_{corr}/T) versus 1000/T at various concentrations of *chamomile extract*.

The activation energy increased in the presence of CE, which indicated physical adsorption (electrostatic) in the first stage [47]. Nevertheless, the adsorption of an organic molecule is not only a physical or chemical adsorption phenomenon. A wide spectrum of conditions ranging from the dominance of chemisorption or electrostatic effects can be seen in other adsorption experimental data [48]. The activation energy rose with increasing inhibitor concentration, suggesting strong adsorption of inhibitor molecules at the metal surface. The increase in the activation energy was due to the corrosion reaction mechanism in which charge transfer was blocked by the adsorption of CE molecules on the mild steel surface [49]. It also revealed that the whole process was controlled by the surface reaction since the energy of the activation corrosion process in both the absence and presence of CE was greater than 20 kJ mol⁻¹ [50].

As seen in Table 4, the values of ΔH_a^* were the same, and the values of E_a increased for the corrosion of mild steel in the presence of inhibitor, indicating that the energy barrier for the corrosion reaction increased in the presence of inhibitor without changing the dissolution mechanism [51]. The entropy of activation, ΔS_a^* , was negative both in the absence and presence of inhibitor, implying that the activated complex represented the rate-determining step with respect to the association rather than the dissociation step. This implies that a decrease in disorder occurred when proceeding from the reactants to the acti-vated complex [52]. In addition, the less negative values of ΔS_a^* in the presence of inhibitor imply that the presence of inhibitor created a near-equilibrium corrosion system state [45].

Inhibitor	C (g/L)	E _a (kJ/mol)	ΔH^*_a (kJ/mol)	ΔS^*_a (J/mol)
1 M HCl	-	33.99	31.21	-100.32
chamomile extract	3.0	38.05	35.46	-93.42
	4.0	45.16	42.51	-70.11
	5.0	45.53	42.93	-81.23
	6.0	55.78	53.18	-36.92
	7.0	56.87	54.27	-36.47

Table 4. The values of activation parameters ΔH_a^* and ΔS_a^* for C38 steel in 1M HCl in the absence and the presence of various concentrations of chamomile extract.

3.2.3 Adsorption isotherm

Adsorption isotherm study describes the adsorptive behaviour of organic inhibitors to know the adsorption mechanism. The most usually used adsorption isotherms are Langmuir, Temkin, Frumkin and other various isotherms. The surface coverage (Θ) values were calculated using polarization measurements.

Langmuir adsorption isotherm were tested and found most appropriate isotherm to explain the experimental data. Langmuir adsorption isotherm is represented by following equations [53]:

$$\frac{\Theta}{1-\Theta} = K_{ads}C\tag{6}$$

The rearrangement gives the following equation:

$$\frac{C}{\Theta} = \frac{1}{K_{ads}} + C \tag{7}$$

Where, K_{ads} is the adsorption equilibrium constant and C is the concentration of inhibitor used in the corrosive medium. A straight line was obtained by plotting the graph of C vs C/ Θ with the R value almost unity (0.9999) (Figure 6). The slope is reported almost unity suggesting that the Langmuir adsorption isotherm model provides the best description of the adsorption behaviour.



Fig . 6. Plots of Langmuir adsorption isotherm of chamomile extract on the steel surface at 298K.

3.3 SEM measurements

Surface morphology of C38 steel was studied by scanning electron microscopy after 6 h immersion in 1 M HCl before and after addition of the inhibitor. Figure 7(a) represent the micrograph obtained of polished steel without being exposed to the corrosive environment while Figure 7(b) showed strongly damaged steel surface due to the formation of corrosion products after immersion in 1 M HCl solution. SEM images of steel surface after immersion in 1 M HCl solution. SEM images of steel surface after immersion in 1 M HCl with 7 g/L CE is shown in Figure 7(c). It can be seen from Fig. 7a that the C38 steel sample before immersion seems smooth and shows some abrading scratches on the surface. Inspection of Fig. 7b reveals that the C38 steel surface after immersion in uninhibited 1 M HCl shows an aggressive attack of the corroding medium on the steel surface. In contrast, in the presence of 7 g/L chamomile extract (Fig. 7c), the steel surface was corroded only negligibly. In addition, there was an adsorbed film on the steel surface that was not observed in Fig. 7b. The result was an enhancement of surface coverage on the steel surface such that there was a decrease in contact between the steel and the aggressive medium. Thus, a good absorptive protection layer can efficiently inhibit the corrosion of steel.



(a) (b) (c) Fig. 7. SEM (x2000) of tinplate (a) before immersion (b) after 6 hours of immersion in 1M HCl (C) after 6 hours of immersion in 1M HCl + 7 g/l of *CE*.

CONCLUSION

The following main conclusions are drawn from the present study:

1. The inhibition efficiency of CE increases with increase in inhibitor concentration.

2. The inhibitor showed maximum inhibition efficiency 88% at 7 g/L concentration.

3. The inhibition efficiency of CE decreased with temperature, which leads to an increase in activation energy of the corrosion process.

4. The negative values of ΔG° ads indicate spontaneous adsorption of the inhibitor on the surface of steel.

5. Langmuir adsorption isotherm and SEM studies showed that CE inhibits through adsorption mechanism.

6. Surface morphological studie with SEM showed that a film of inhibitor is formed on the electrode surface.

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