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Short Communication

The Anti-Corrosion Behavior of *Lavandula dentata* Aqueous Extract on Mild Steel in 1M HCl

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The corrosion inhibition of mild steel in 1 M hydrochloric acid solutions by aqueous extract of *Lavandula dentata* (*LD*) was investigated using chemical (weight loss) and electrochemical (potentiodynamic and electrochemical impedance spectroscopy, EIS) measurements. The results from these techniques show that the inhibition efficiency increases with increase in the percentage of LD extract to attain 95% at 2wt%. Polarization studies indicate that the phytochemical compounds present in LD extract acts as a mixed type inhibitor in 1 M HCl solution.

Keywords: *Lavandula dentata* ; corrosion; inhibition; Mild steel.

1. INTRODUCTION

Corrosion associated to the use of acid solutions in industrial applications [pickling, cleaning, descaling, ….] cost to industrial sector billions of dollars each year for preventing and replacement of maintenance [1,2]. To reduce the aggressivity of acidic mediums against structural materiel such as mild steel, corrosion inhibitors are applied.

Most of the efficient acid inhibitors are organic compounds containing nitrogen, sulphur and/or oxygen atoms in their molecules [**Error! Bookmark not defined.**-5]. Nevertheless, these compounds are in general expensive, toxic and not biodegradable.

The use of the natural products such as extracted compounds from leaves or seeds as corrosion inhibitors have been widely reported. Shauhan and Gunasekaran [6] studied the corrosion inhibition of steel by eco-friendly *Zenthoxylum alatum* plant extract in HCl as well as in phosphoric acid medium. Essential oils and leaves extracts are used as common corrosion inhibitors. The anticorrosion activity of leaves extracts as *henna* [7], *Artemisia* [8-10], *pepper* [11], *lavender* [12], *and Accacia conicianna* [13] was investigated. Corrosion inhibition has also been studied for the essential oils and extracts *of lawsonia* [14]. Similar results were also shown by *Rosmarinous officinalis* [15], *Annonas quamosa* [16], *Acacia Arabica* [17], *Carica papaya* [18], *Azadirachta indica and Vernonia amydalina* which were used for steel in acid media. *Nypafructicans wurmb* leaves were studied for the corrosion inhibition of mild steel in HCl media [19-21].

The use of plant extracts as corrosion inhibitors are justified by the phytochemical compounds present therein with molecular structures bearing an effective corrosion inhibition groups such as hydroxyl [OH], carbonyl [CO], NH, aromatic ring and so [22, 23] by which these compounds can adsorb on metal surface and block the active surface sites to reduce corrosion [24].

The aim of the present work is to evaluate the inhibitive effect of aqueous extract of LD extract as a green corrosion inhibitor on the corrosion of mild steel in 1M HCl solution. The assessment of the corrosion behavior was studied using weight loss, potentiodynamic polarization measurement, and electrochemical impedance spectroscopy [EIS].

2. MATERIALS AND METHODS

2.1. Materials

The chemical composition of the mild steel that was used in this woek is C 0.01%, P 0.03%, S 0.03 %, Mn 0.85%, Fe 99.08%. Analytical grade 37% HCl was purchased from SOMAPROL [Morroco]. Aerial parts of Lavandula dentata were collected at the flowering stage in April 2011 from Tafoughalt [Oujda region, Northeast of Morocco].

2.2. Preparation of plant extact

Aerial parts of *Lavandula dentata* were air-dried at room temperature and essential oil was discarded by hydrodistillation for 3 h using a Clevenger-type apparatus. The aqueous extract was centrifuged at 1000rd/min for 10 min and filtered to eliminate the undissolved materials. The aqueous solution obtained was concentrated by vacuum evaporation and dried with a sand bath to constant weight before use. For corrosion tests the weighted portions of the dried sample were used.

2.3. Preparation of specimens

Mild steel pieces of lxbt [1,5x1,5x0,2 cm]were polished successively using 800, 1000 and 1200 gritted emery paper. Next, it were washed with double distilled water and dried prior to use.

2.4. Corrosion tests

2.4.1. Weight loss measurements

The weights of specimens were noted and then they were immersed in 1M HCl solution [50 ml] containing various concentrations of LD extract. After the duration of 4h, The specimens were removed from the acidic solution, washed with distilled water, dried and finally weighed. The differences in weights were noted and the inhibition efficiencies [E_w %] were calculated using the following equation (eq. 1) :

$$E_W\% = \frac{W_{corr} - W'_{corr}}{W_{corr}} \times 100 \qquad (1)$$

where W_{corr} and W'_{corr} are the values of the weight loss in [g] of mild steel in the absence and presence of corrosion inhibitor respectively.

2.4.2. Potentiodynamic measurements

Polarization measurements were performed in a thermally jacketed conventional three-electrode Pyrex cell with a platinum foil counter electrode and saturated calomel electrode [SCE] coupled to a fine Luggin capillary as the reference electrode. All measurements were carried out using on an electrochemical measurement system (Volta- Lab 40) comprised of a PGZ 100 potentiostat, a PC and Voltamaster 5.3 electrochemical software.

The potentials were recorded at a scan rate of 1 mV/s from the corrosion potential [E_{corr}] in the cathodic direction and subsequently in the anodic direction. The electrode was held in the test environment for 30 min prior to each experiment, which proved sufficient for E_{corr} to attain a reliable stable value.

Corrosion current density values were obtained by Tafel extrapolation method.

Inhibition (E_I%) as function inhibitors concentrations, E_I% was calculated according to the following equation (eq. 2) :

$$E_I\% = \frac{I_{corr} - I'_{corr}}{I_{corr}} \times 100 \qquad (2)$$

where I_{corr} and I'_{corr} are the uninhibited and inhibited corrosion current densities respectively.

2.4.3. Electrochemical impedance measurements

Experimental impedance spectra were obtained in the frequency range of 100 KHz to 10 mHz with ten points per decade at the corrosion potential. A sine wave with 100 mV amplitude was used to perturb the system. The Nyquist diagrams obtained were automatically controlled by computer programs.

The percent inhibition efficiency, E_R (%), is calculated by polarization resistance obtained from Nyquist plots, according to the equation [eq. 3]:

$$E_R\% = \frac{R_t^{-1} - R_{t(inh)}^{-1}}{R_t^{-1}} \times 100 \qquad (3)$$

where $R_{t[inh]}$ and R_t are the polarization resistance values with and without inhibitor respectively.

3. RESULTS AND DISCUSSION

3.1. Gravimetric measurements

Although there are many experimental techniques which can be used to evaluate the inhibitor efficiency of LD extract, weight loss is one of the simplest and frequently used methods.

Using the experimental weight loss data, percentage inhibition efficiencies (E_W %), were calculated in the classical way. Values of E_W for all investigated percentages of LD extract in 1 M HCl are summarized in Table 1.

Table 1. Inhibitor efficiencies for various percentages of *Lavandula dentata* extract [LD] for the corrosion of mild steel in 1M HCl from weight loss measurements.

Percentage of LD (wt %)	W(mg/cm^2.h)	E_W (%)
-	0.473	-
0.5	0.115	75
1	0.091	81
1,5	0.051	89
2	0.024	95

A measurement for different percentages of each inhibitor inhibits the corrosion of the steel sample. The corrosion rate values decrease when the inhibitor percentage increases due to the increase of the inhibition efficiencies. At this purpose, one observes that the optimum percentage of inhibitor required to achieve the efficiency is found to be 2% (E_w = 95%).

Adsorption isotherms are very important in determining the mechanism of organic electrochemical reactions. The most frequently used adsorption isotherms are Langmuir, Temkin and Frumkin. So several adsorption isotherms were tested for the description of adsorption behaviour of studied compound and it is found that adsorption of compound under study obeys the Langmuir adsorption isotherm (Fig. 1). The experimental result is in good agreement with the Langmuir adsorption isotherm, which is represented by the following equation (eq. 4):

$$\frac{C_{inh}}{\theta} = \frac{1}{K_{ads}} + C_{inh} \qquad (4)$$

where C_{inh} , θ and K_{ads} are the inhibitor concentration, the surface coverage calculated by E%/100 and the adsorption equilibrium constant respectyvely.

The plot of C/θ versus C yields straight line with slope of almost equal unity (regression coefficient is 0.998). The value of K_{ads} is found to be 972.47 dm^3 mol^{-1}.

We limited to this stage and the value of $\Delta G°_{ads}$ was not determined because of the infinite components of natural extract of Lav [25-28].

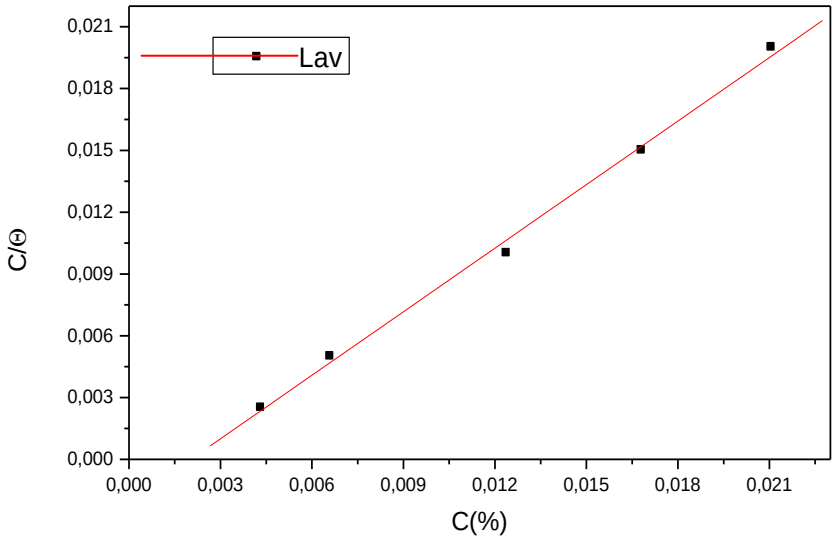

Figure 1. Langmuir's isotherm for adsorption of LD compounds on the surface of the mild steel in 1 M HCl.

3.2. Potentiodynamic measurements

Figure 2 represent the potentiodynamic polarization curves of mild steel in 1 M HCl in the absence and presence of various percentages of the LD extract compounds under study.

The values of corrosion potential (E_{corr}), anodic and cathodic Tafel slopes (βa and βc), corrosion current density (I_{corr}) and percentage inhibition efficiency (E_I %) are presented in Table 2.

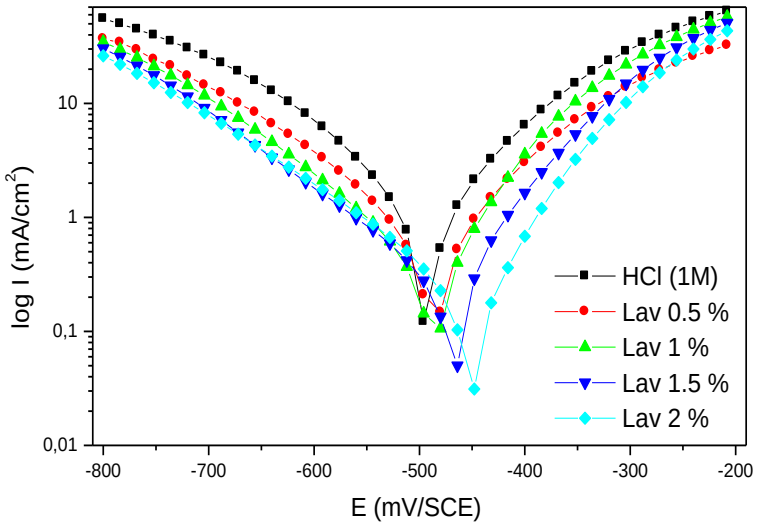

Figure 2. Potentiodynamic polarisation curves of stainless steel in hydrochloric acid solution with and without inhibitor at different percentages of LD extract.

Table 2. Potentiodynamic polarisation parameters of the stainless steel in HCl solution without and with addition of various LD extract percentages at 35°C.

Percentages of LD extract	E_{corr} (mV/ECS)	$-\beta_C$ (mV/dec)	β_a (mV/dec)	I_{corr} ($\mu A/cm^2$)	E_I (%)
-	-543	257.4	220	450.8	-
0.5	-540	260.1	221	228.2	49
1	-556	189.4	154.8	150.4	67
1,5	547	166.4	135.9	86.7	81
2	-535	172.3	133.6	73.7	84

From these results we can conclude that the corrosion densities calculated by Tafel extrapolation decreased with increasing of inhibitor concentrations. This behaviour reflects that the compounds of LD extract are adsorbed on the anodic sites and hence inhibition occurs. This means the inhibitor under investigation acts as a mixed type inhibitor. The corrosion potential of the inhibitor containing solution remains almost unchanged to that in the solution without the inhibitors. The slopes of the Tafel line decreases indicating that the inhibition effect is caused by adsorbed inhibiting species [29-32].

3.3. The Impedance spectroscopy

Impedance diagram was plotted to obtain more details on the mechanism of the inhibitor action (Fig.3). The Nyquist plot obtained in the absence and in the presence of the inhibitor showed a semi-circle corresponding to a phenomenon of charge transfer.

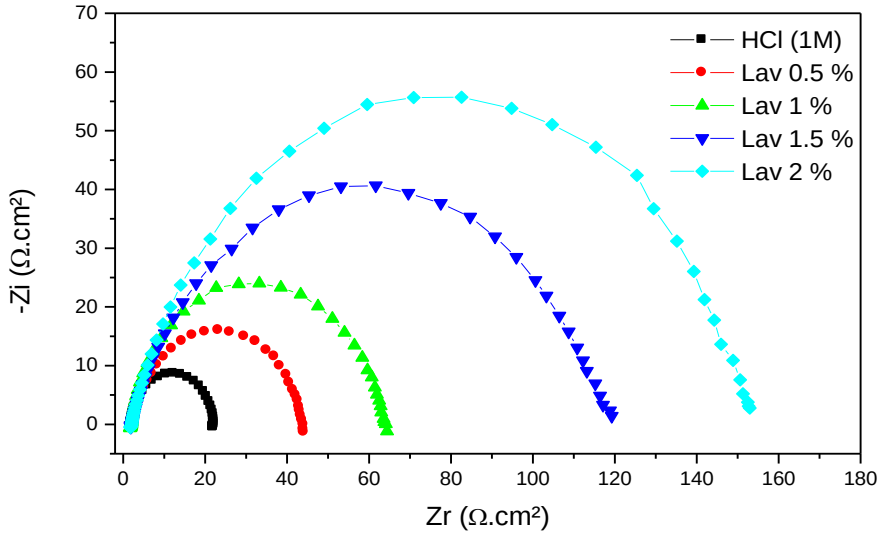

Figure 3. Impedance plot of mild steel obtained in 1M HCl in the absence and presence of various concentrations of LD extract.

Various parameters such as polarization resistance (R_t), double layer capacitance C_{dl} and percentage inhibition efficiency [E_R %] have been calculated and listed in Table 3. The polarization resistance values have been calculated from the difference in impedance at higher and lower frequencies [28]. The values of C_{dl} were obtained at f_{max} using the following equation:

$$C_{dl} = \frac{1}{2\pi f_{max}} \frac{1}{R_t} \qquad (6)$$

Table 3. Impedance parameters for corrosion of mild steel in hydrochloric acid containing different percentage of LD extract.

Percentage of LD [%]	R_s (Ω.cm²)	R_t (Ω.cm²)	f_{max} (Hz)	C_{dl} (μF/cm²)	E_R (%)
-	2.272	20.49	86.55	978.2	-
0.5	2.217	42.25	63.29	59.5	51%
1	1.970	59.76	31.64	84.2	65%
1.5	2.131	115.2	15.82	87.3	82%
2	2.037	151.5	10	105	86%

From Table 3, it was clear that charge transfer resistance R_t values were increased and the capacitance values C_{dl} decreased with increasing inhibitors' concentration. The decrease in the capacitance which can result from a decrease in local dielectric constant and/or an increase in the thickness of the electrical double layer, suggests that the inhibitor molecules act by adsorption at the metal/solution interface.

The presence of LD extract compounds increases the impedance but does not change other aspects of the behaviour. These results support those of polarization measurements that the inhibitor does not alter the electrochemical reactions responsible for corrosion. It inhibits corrosion primarily through its adsorption on the mild steel surface. The inhibition effect may be due to the various components in extract. The work of Upson et al. [33] in 2000 on *Lavandula dentata* cultivated from Agadir and Alhoceima regions, showed the presence of the flavone C-glycoside, vitexin, 6-hydroxyflavone glycosides. This kind of molecules having several active centers may play major role in adsorption phenomenon.

The results obtained from EIS measurements are in good agreement with that obtained from potentiodynamic polarization and weight loss measurements.

5. CONCLUSION

The LD extract compounds inhibit effectively the corrosion of mild steel in 1 M HCl. Indeed, the optimum percentage of inhibitor required to achieve the efficiency is found to be 2% (E_w = 95%).

The polarization curves indicate that the extract of LD act as mixed type inhibitor and the impedance data indicate that inhibition was achieved via adsorption. The adsorption of LD extract is well described by Langmuir isotherm model.

Double layer capacitances decreases with respect to the blank solution when LD extract is added. This fact may be explained on the basis of adsorption of LD extract compounds on the steel surface.

In determining the corrosion, electrochemical studies and weight loss measurements give similar results.

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