Salvia officinalis essential oil as green corrosion inhibitor for mild steel in acidic media

S. RHAIMI[1], M. OUAKKI[2,4*], N. Rhaiem[1], M. GALAI[3], K. Dahmani[2], F. EL FARHANI[3], M. BARRAHI[1], M. Ebn Touhami[3], M. OUHSSINE[1]

[1]*Laboratory of Biodiversity and Natural Resources, Faculty of Sciences, Ibn Tofail University, Kenitra, Morocco.*

[2]*Laboratory of Organic Chemistry, catalysis and Environment, Faculty of Sciences, Ibn Tofaïl University, PO Box 133, 14000, Kenitra, Morocco.*

[3]*Laboratory of Advanced Materials and Process Engineering, Faculty of Sciences, University Ibn Tofail PB. 133-14000, Kenitra, Morocco*

[4]*National Higher School of Chemistry (NHSC), University Ibn Tofail BP. 133-14000, Kenitra, Morocco*

Abstract

The main aim of this research is to study the inhibition effect of Salvia officinalis against the corrosion of mild steel in 1 M HCl solution utilizing Potentiodynamic Polarization (PDP) and Electrochemical Impedance Spectroscopy (EIS), in the presence of various concentrations of the examined *Salvia officinalis* essential oil (EO). PDP findings show that the examined EO behaved as a mixed type inhibitors with a predominant cathodic. EIS measurements indicated that the EO could inhibit the corrosion of mild steel by the formation of a protective film on the surface of mild steel. The inhibition efficiency of Salvia officinalis followed the order: 81.2% at concentration of 2.0g/l.

Keywords: Corrosion inhibition, *Salvia officinalis,* essential oil, mild steel.

1. Introduction

Corrosion is the destructive attacks of metals by its environment. The spontaneous destruction of metal due to heterogeneous chemical reaction is the chemical corrosion [1]. The serious consequences of the corrosion process have become a problem of worldwide significance. In addition to our everyday encounters with this form of degradation, corrosion causes enormous economic damage. Millions of dollars are lost each year because of corrosion [2]. Acid solutions such as HCl and H_2SO_4 are widely used in industry: some of the important fields of application are acid pickling of steel, chemical cleaning and processing, ore production and oil well acidizing. A major drawback is the susceptibility of iron to corrosion [2-4]. The combat against corrosion is has been undertaken by many scientists and engineers. The best way to protect materials against corrosion is using inhibitors [5, 6]. This use, even in small concentrations has a significant decrease in the percentage of corrosion on the surface of the metal [7, 8]. One of the most widely used practices for this is the use of corrosion inhibitors, both organic and inorganic. Use of many

inorganic inhibitors, particularly those containing phosphate, chromate and other heavy metals are now being gradually restricted by various environmental regulations [9]. Therefore, the study of new non-toxic corrosion inhibitors is essential to overcome this problem. Inhibitors in this class are those that are environmentally friendly and are obtained from natural products such as plant extracts. Recently, several studies have been carried out on the inhibition of corrosion of metals by plant extract and essential oils [10, 11]. In this context, our laboratory chose to test essential oil of *Salvia officinalis* as a green corrosion inhibitor for mild steel in acidic media.

This work aims to evaluate the inhibitory efficacy of the essential oil of *Salvia officinalis* on mild steel. The intrinsic corrosion protective properties of this inhibitor on mild steel were investigated using electrochemical impedance spectroscopy (EIS) and potentiodynamic

polarization measurements. This study also aimed to predict the thermodynamic feasibility of *Salvia officinalis* on metallic surfaces. Additionally, detailed investigations of the effect of temperature on the electrochemical parameters of the system were also studied and discussed to improve the understanding of the adsorption mechanism of the studied inhibitor.

2. Materials and methods

2.1. Plant material

The aerial parts of *S. officinalis* were harvested in the wild of Kenitra (Morocco). A voucher specimen was deposited in drying oven. The dried plant material was stored at room temperature (298 K) and in the shade before the extraction.

2.2. Extract of Salvia officinalis essential oil

The extraction of essential oil from the aerial parts of *S. officinalis* was conducted by hydrodistillation using a Clevenger type apparatus (Clevenger, 1928), and the essential oil yield was 1%. The essential oil obtained was dried and stored at 277 K in the dark before analysis.

2.3. Solutions and steel samples preparation

The studies were carried out on mild steel samples, composed of 0.17 wt% carbon C, 0.37 wt% manganese Mn, 0.20 wt% silicon Si, 0.03 wt% sulfur S, 0.01 wt% phosphorus P and balance Fe. Before each measurement, the steel samples were prepared by polishing with emery paper of increasingly fine grain size (from 180 to 2000), followed by cleaning with distilled water and ethanol, and finally drying at room temperature. All experiments were performed in an aggressive 1.0 M HCl solution. This aggressive solution was prepared by diluting analytical grade HCl (37 wt. %) with distilled water. The concentration range of the essential oil studied in this work was set from 0.5g/l to 2.0 g/l. This concentration range was determined after studying the solubility of the inhibitors in the corrosive medium, and the blank solution was also prepared for comparison.

2.4. Electrochemical measurements

a. Potentiodynamic polarization

The assembly used was composed of three electrodes (a platinum counter electrode, *Saturated calomel electrode* (*SCE*) and a working electrode (WE)) supplemented with a computer-controlled potentiostat PGZ100, VOLTALAB type and « VoltaMaster4 software. The electrode was maintained at 30 min of immersion at the corrosion potential. Before starting tracing of the curves (cathodic and anodic), the working electrode was maintained at its abandonment potential for one hour. The intensity of the current was measured between the working electrode and the platinum counter-electrode. The plotting of the curves (intensity-potential) allowed the determination of the corrosion potential (E$_{corr}$), the corrosion current density (i$_{corr}$), *anodic*(βa) and *cathodic*(βc) *Tafel slopes*. The

inhibitive effectiveness of corrosion E (%) is defined by following equation:

$$\eta_{PDP}\% = \left[\left(i_{corr}^{\circ} - i_{corr} \right) \middle/ i_{corr}^{\circ} \right] \times 100$$

i$^0_{corr}$ and i$_{corr}$ are respectively values of density of the corrosion current with and without inhibitor, they are determined by extrapolation of the cathode and anode lines of Tafel to the potential of corrosion [12].

b. Electrochemical impedance spectroscopy

The electrochemical impedance spectroscopy measurements were carried out using a transfer function analyzer (VoltaLab PGZ 100), with a small amplitude a.c. signal (10 mV rms), over a frequency domain from 100 kHz to 100 mHz with five points per decade. The EIS diagrams were done in the Nyquist representation. The results were then analyzed in terms of an equivalent electrical circuit.
The inhibiting efficiency derived from EIS, η_{EIS}% was also added and calculated using the following equation:

$$\eta_{imp}\% = \frac{(R_{ct} - R_{ct}^{\circ})}{R_{ct}} \times 100 \theta = \frac{(R_{ct} - R_{ct}^{\circ})}{R_{ct}}$$

With R$^0_{ct}$ and R$_{ct}$ indicated the charge transfer resistances in the nonexistence and existence of inhibitor, respectively, and θ was the recovery rate.

3. Results and discussion

3.1. Potentiodynamic polarization (PP)

PP technique is one of the electrochemical techniques that provides information on the kinetics of the corrosion process at the metal-solution interface. The polarization curves of Mild Steel in the investigated medium in both absence and presence of different concentrations of the essential oil of *Salvia officinalis* at 298 K is depicted in Fig.1. It can be noticed form Fig. 1. that only the cathodic regions show a wide range of linearity, which reveals that the Tafel's law is well verified in the cathodic domain and the hydrogen reduction controlled by pure activation kinetics[1], hence the decrease of the current densities of the cathodic and anodic branches, results in the reduction of the anodic dissolution of the steel, and consequently the blocking of the hydrogen evolution reaction [13], These findings can be attributed to the adsorption of essential oil of *Salvia officinalis* on the mild steel surface as a barrier that prevents the corrosion process [14]. The electrochemical parameters such as the corrosion current densities (i$_{corr}$), the corrosion potentials (E$_{corr}$), the cathodic slopes (βc), anodic slopes (βa), and percentage inhibition efficiency (η$_{PP}$%) for the different concentrations of essential oil of *Salvia officinalis* are shown in Table 1. The electrochemical parameters deduced from the potentiodynamic polarization curves (Table 1) show that the corrosion current density (i$_{corr}$) decreases with the increase of the concentration of the essential oil of *Salvia officinalis* plant to reach a minimum value of 171 μA cm^{-2} in the presence of 2.0 g/l (optimal concentration). As a result, the inhibition efficiency increases and reaches 82.6 %. This increase suggests that the inhibitor molecules form an adsorbed layer on the mild steel surface that blocks the corrosion process, and its effect is

progressively increased by increasing the inhibitor concentration.

The existence of essential oil of *Salvia officinalis* caused a change in the values of E_{corr} compared to the blank solution. According to the literature [15, 16], if the shift of E_{corr} with inhibitor is higher than $\pm$ 85 mV compared to the uninhibited case, the inhibitor may be considered as a cathodic or anodic type inhibitor. On the other hand, if the E_{corr} variation is less than $\pm$ 85 mV, the corrosion inhibitor can be considered as a mixed type inhibitor. In this investigation, the potential variation does not exceed +85 mV, which indicated that these essential oils were mixed type inhibitors with a predominant inhibition influence on the cathodic reaction.

3.2. Electrochemical impedance spectroscopy

Electrochemical Impedance Spectroscopy (EIS) is particularly suitable technique for understanding the mechanism of action of inhibitor. In addition, this technique allows evaluating the dielectric characteristics of the formed film and following their evolution according to many parameters. It also makes it possible to explain the chemical or electrochemical processes developing through the formed films [17,18]. Thus, in order to complete and compare the results obtained by the potentiodynamic technique, the corrosion inhibition of mild steel in the 1.0 M HCl medium in the absence and presence of essential oil of *Salvia officinalis* was investigated by electrochemical impedance spectroscopy (EIS). Fig. 2 presents the impedance diagrams in the Nyquist plan for the mild steel after 0.5h of immersion in the corrosive medium before and after adding in the essential oil at 298K. The Nyquist diagrams represent single capacitive loop that is not a perfect semicircle, which is attributed to the frequency dispersion of the interfacial impedance. The existence of a single capacitive loop reflects the presence of a system that follows pure charge transfer kinetics [13, 14, 15]. Moreover, the increase in loop size as inhibitor concentrations increase characterizes the increase in the protective capacity of the inhibitor at the metal-solution interface [19]. EIS spectra could be assimilated, by analogy, to electrical impedance. The different processes that occurs at the electrode/electrolyte interface can be modelled using an equivalent electrical circuit which is depicted Fig. 3. This circuit contains a charge transfer resistance (R_{ct}), solution resistance (Rs) and a constant phase element (CPE). It was necessary to introduce CPE in the circuit to explain the depression of the capacity semicircle corresponding to the surface heterogeneity resulting from the surface roughness, adsorption of inhibitors and impurities [20]. The impedance of this element is frequency-dependent and can be calculated using the equation [10].

$$Z = \frac{1}{Q(j\omega)^n}$$

Q is the CPE constant (in $\Omega^{-1} \cdot Sn \cdot cm^{-2}$), ω is the angular frequency (in $rad \cdot s^{-1}$), $j^2 = -1$ is the imaginary number and n is a CPE exponent which can be used as a gauge for the heterogeneity or roughness of the surface [6,21]. The double layer capacitances, C_{dl}, for a circuit including a CPE were calculated by using the following Equation:

$$C_{dl} = \sqrt{Q \times R_{ct}^{1-n}}$$

The Electrochemical parameters determined from impedance plots are listed in Table 2.

As shown in Table 2, the charge transfer resistance (R_{ct}) becomes higher within creasing concentration of essential oil in the examined corrosive solution. On the other hand, the decrease in the value (C_{dl}) in the presence of the studied essential oil to the blank solution can be attributed to the low local dielectric constant, resulting from the progressive replacement of water molecules by the adsorption of essential oil of *Salvia officialinalis* at the interface of the mild steel. In addition, the values of the factor "n" in the presence of the studied essential oil were higher than the uninhibited medium, which shows that the homogeneity of the Mild Steel is reduced due to the formation of an organic film on the steel surface [19]. The inhibitory efficiency increases with increasing essential oil concentration and reaches an optimum value of 81.2 % at 2.0 g/l, similar trends were observed from the polarization representation, confirming the same results and the same corrosion inhibition process.

3.3. Effect of temperature

Temperature is one of the factors that can modify both the behavior of a metal in a corrosive environment and would favor the desorption of the inhibitor, as well as a rapid dissolution of the organic compounds or complexes formed. The influence of temperature on the inhibitory efficacy of Salvia officinalis essential oil was also studied by potentiometer. The polarization curves obtained in 1 M acid medium (HCl) with and without the addition of 2.0 g/l essential oil of *Salvia officinalis* at a temperature range from 298K to 328K, are presented in figure 4. For the characteristic parameters of these curves, they are illustrated in table 3. According to Fig. 4, it can be seen that the increase in temperature leads to an increase in the anodic and cathodic current densities in the absence and presence of the studied essential oil. This indicates the positive evolution of the corrosion kinetics of mild steel in the presence of the inhibitor. Moreover, it can be noticed that the curves are more or less parallel to each other, which indicates that the temperature only affects the corrosion rate and does not influence the corrosion mechanism. The results listed in table 3 reveals that the value of corrosion current density (i_{corr}) increases with increasing temperature in the absence and the presence of the essential oil. This result indicates an increase in the corrosion rate of mild steel at the same time the acceleration of the metal dissolution process. Moreover, the inhibition efficiencies values decrease slightly with increasing temperature and reach 76.4% at 328K which indicates the weakening of the protective film [22].

3.4. Activation parameters of corrosion process

The activation thermodynamic parameters of the corrosion process can be determined using Arrhenius Eq. and Eq. transition state [23].

$$i_{corr} = Ae^{\left(-\frac{E_a}{RT}\right)}$$

$$\ln\left(\frac{i_{corr}}{T}\right) = \left[\ln\left(\frac{R}{hN_a}\right) + \left(\frac{\Delta S_a}{R}\right)\right] - \frac{\Delta H_a}{RT}$$

Where Ea is the apparent activation energy, R is the gas constant; A is the Arrhenius pre-exponential factor, N is Avogadro's number, h is Plank's constant, ΔSa is the entropy of activation and ΔHa is the enthalpy of activation.

A plot of ln (i$_{corr}$) vs 1000/T obtained gave a straight line with regression coefficient close to unity, as shown in Figure 5. The Arrhenius plot of ln (i$_{corr}$/T) vs 1000/T (Figure 5) which gave straight lines with slope ΔHa/R and intercept (ln (R/N.h) + ΔSa/R) from which ΔHa and ΔSa values were

calculated. The values of ΔHa and ΔSa were calculated and listed inTable 4. It was observed that the apparent activation energy value Ea is higher in the presence of inhibitor than in its absence. This result indicates that the energy barrier for the corrosion reaction increasing in the presence of inhibitor is associated with physical adsorption or weak chemical bonding between and the essential oil [24]. The positive values of ΔHa mean that the dissolution reaction is an endothermic process indicating difficulty of mild steel dissolution [25, 26]. The entropy ΔSa increases negatively in the presence of *Salvia officinalis* essential oil which reflects that the activated complex represents an association rather than a dissociation step which means the formation of an ordered, stable layer of these inhibitors on the mild steel [27].
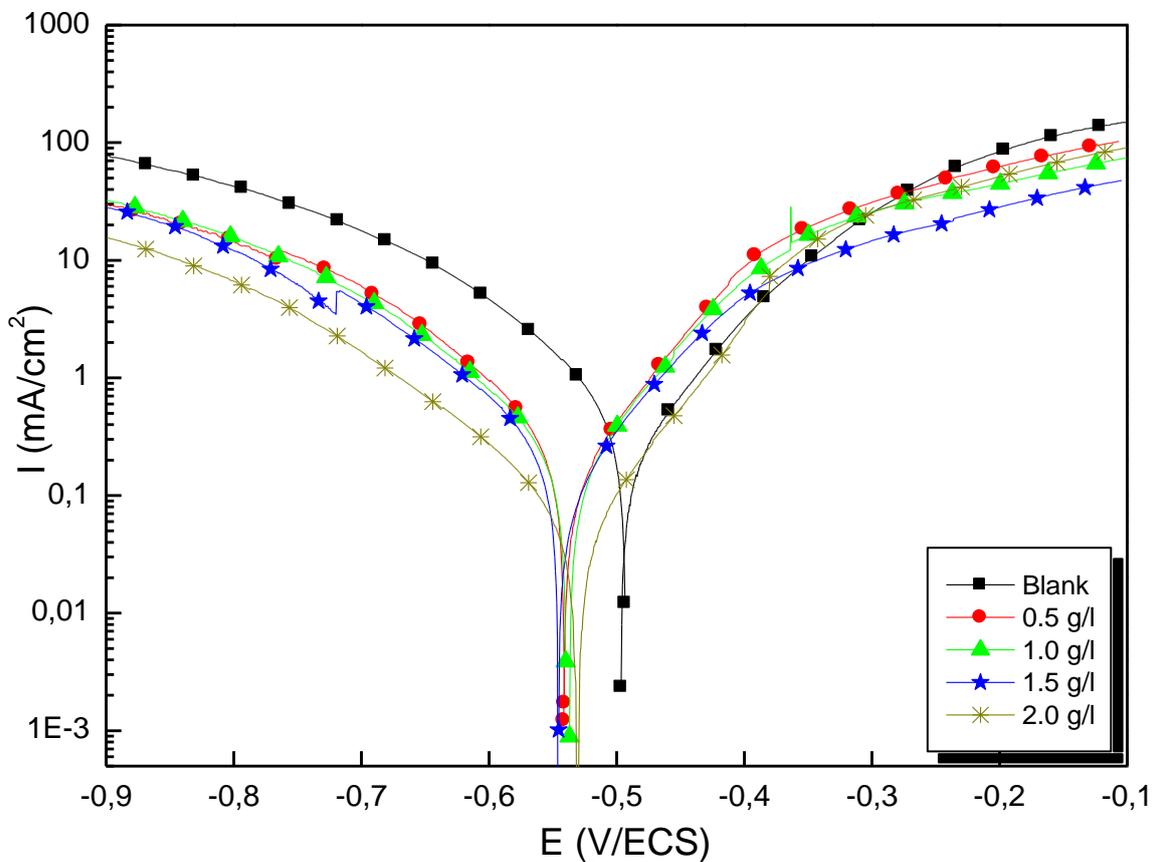

Fig. 1. Polarization curves for Mild Steel in 1.0 M HCl with various concentrations of essential oil of *Salvia officinalis* at 298 K.

Table 1. PDP indices for Mild Steel in 1.0 M HCl with and without essential oil of *Salvia officinalis* at 298 K.

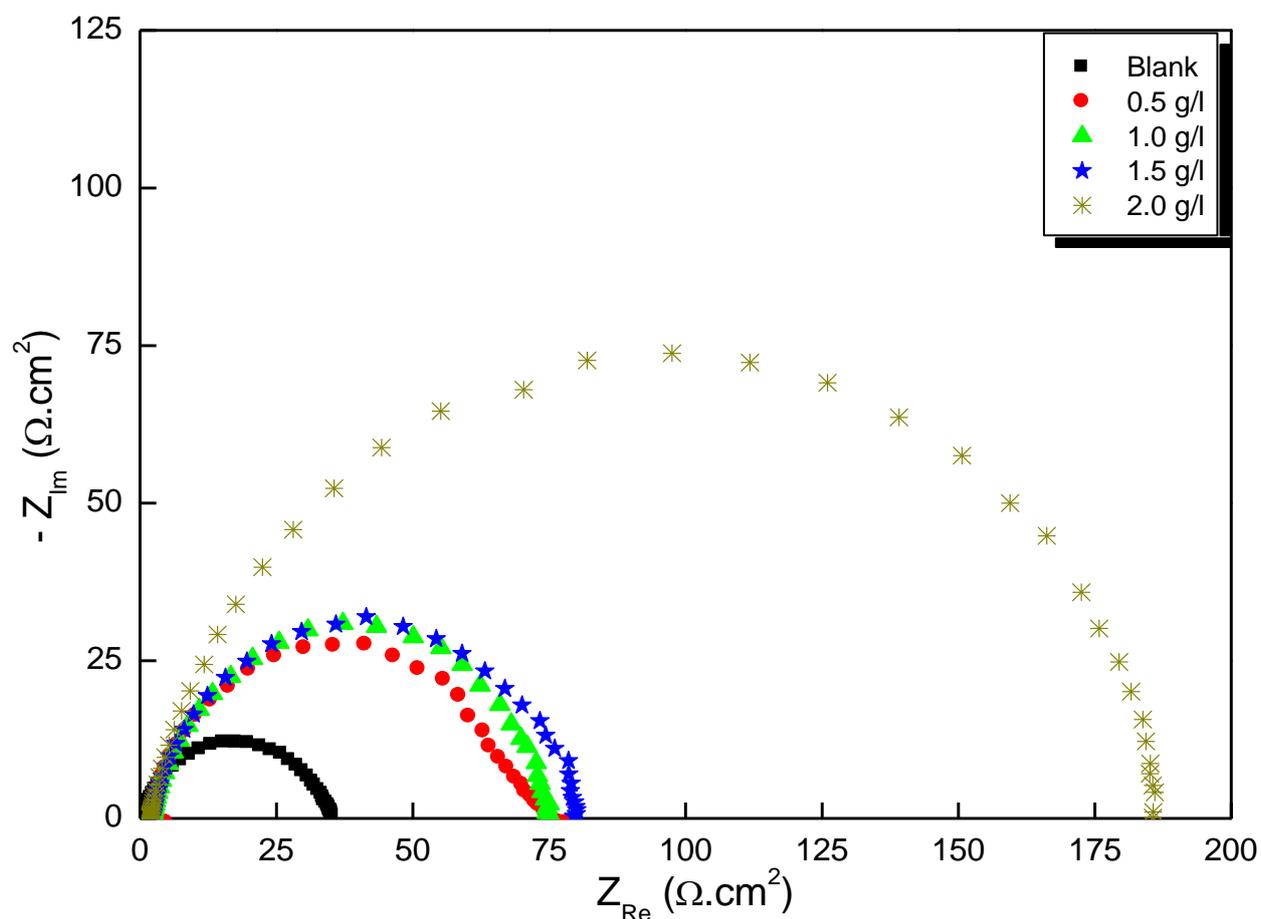| Medium	[E O] (g/l)	E_{corr} (mV/SCE)	i_{corr} ($\mu A\ cm^{-2}$)	$-\beta_c$ (mV dec^{-1})	β_a (mV dec^{-1})	η_{PP} (%)
HCl(1M)	--	- 498	983	140	150	-
E.O	0.5	-538	482	131	120	50,9
	1.0	-537	460	129	121	53,2
	1.5	-545	427	115	122	56,5
	2.0	-529	171	134	118	82,6

Fig. 2. Impedance diagrams in Nyquist representation of steel in HCl solution (1 M) at different concentrations of essential oil of *Salvia officinalis*

Table 2. Electrochemical parameters and inhibitory efficacy of corrosion of mild steel in HCl 1M with and without the addition of essential oil of *Salvia officinalis.*

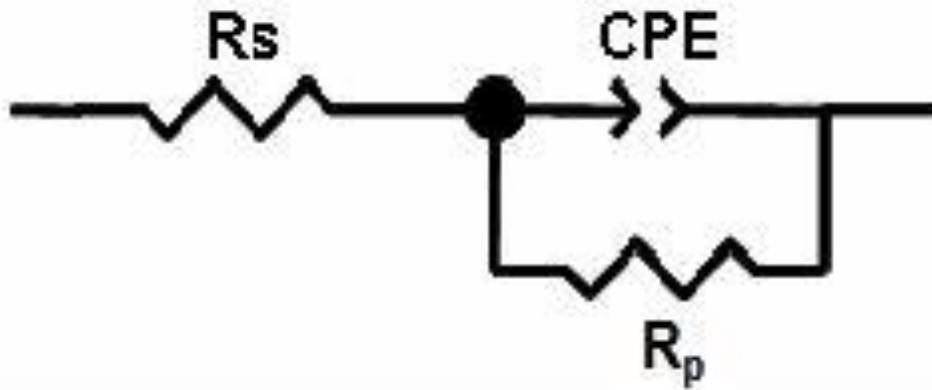|	Conc. (g/l)	R_s (Ω cm^2)	R_{ct} (Ω cm^2)	C_{dl} (μF cm^{-2})	n_{dl}	Q ($\mu F.S^{n-1}$)	Θ	η_{imp}%
HCl 1.0M	--	1.12	34.7	121	0.773	419	-	-
E.O	0.5	2.1	70.44	71.1	0.848	145	0.507	50,7
	1.0	2.1	72.33	68.6	0.873	141	0.520	52,0
	1.5	2.1	78.09	59.4	0.865	104	0.555	55,5
	2.0	1.6	184.9	42.7	0.863	95	0.812	81,2

E.O: Essential oil

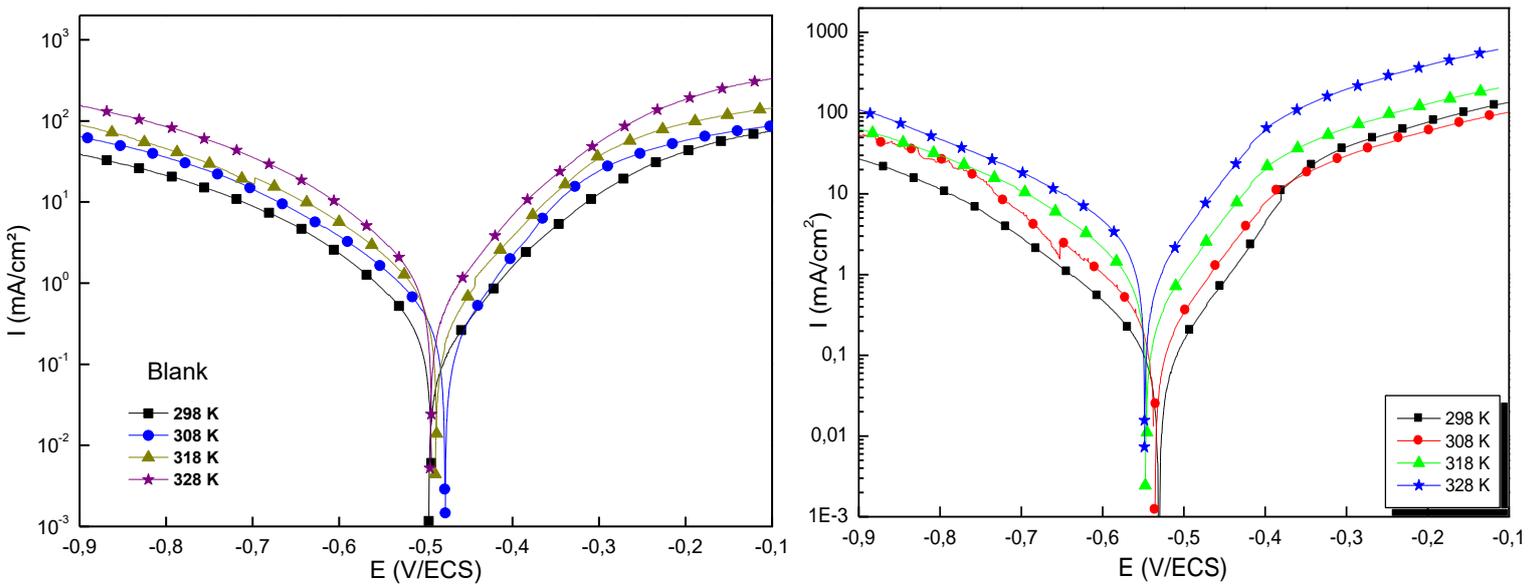**Fig. 3.** Equivalent electrical circuit.

Fig. 4. Polarization curves of steel in 1M HCl medium and at different temperatures in the absence and the presence of essential oil of *Salvia officinalis*

Table 3. Study of the electrochemical parameters and the inhibitory efficiency of E.O against steel in 1 M HCl

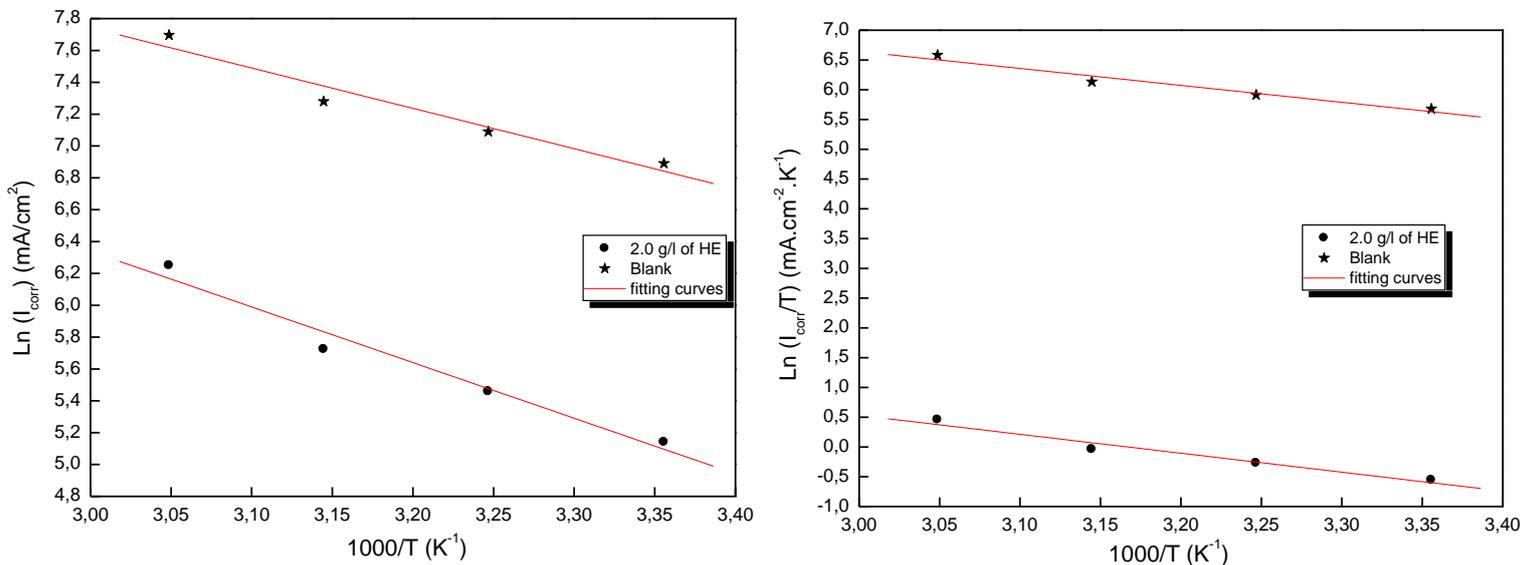| Compounds	Tempe K	E_{corr} mV/SCE	i_{corr} μA cm^{-2}	$-\beta_c$ mV dec^{-1}	β_a mV dec^{-1}	$\eta_{PP}\%$
Blank	298	-498	983	140	150	-
	308	-491	1200	184	112	-
	318	-475	1450	171	124	-
	328	-465	2200	161	118	-
2.0 g/l of EO	298	-529	171	134	118	82,6
	308	-535	235	130	112	80,4
	318	-543	306	135	110	78,8
	328	-546	518	127	106	76,4

Fig. 5. Arrhenius curves of mild steel in 1 M HCl with and without 2.0 g/l of essential oil of *Salvia officinalis*

Table 4. The value of the activation parameters Ea, ΔHa and ΔSa of mild steel in 1M HCl in the absence and in the presence of 2.0 g/l of essential oil of *Salvia officinalis*

Medium	Conc	Ea (KJ/mol)	ΔHa (KJ/mol)	ΔSa (J/mol.K)
1M HCl	Blank	21	18.5	-126
	2.0 g/l HE	29	26.4	-113.7

There was a tendency to increase calcium level (– 15.64%) compared to the control. Based on the above, it can be concluded that the inclusion of various doses of Cellobacterin-T in the diet does not significantly affect the morphological and biochemical parameters of the blood of experimental chickens.

4. Conclusions

All of the above results indicated that Salvia officinalis essential oil is a good corrosion inhibitor for mild steel in an acidic environment (HCl). Gravimetric tests have shown that the effectiveness of the inhibitor increases with the concentration of essential oil and decreases with temperature. The corrosion current density and passivation values are reduced by an order of magnitude when the inhibitor is added, so it is an anodic inhibitor. The essential oil of *Salvia officinalis* found on the surface of the steel begins to be desorbed as a function of time, which decreases its inhibitory effectiveness.

Conflict of interest

The authors declare there are no competing interests regarding the publication of this article.

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