

Full Paper

New Formulation based on *Ceratonia Siliqua L* Seed Oil, as a Green Corrosion Inhibitor of Iron in Acidic Medium

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Abstract- In this study, the effect of the formulation derived from *Ceratonia siliqua L*, (noted FCSL) as green corrosion inhibitor of iron in solution simulated to the rain water (at pH=3.6) was examined by using the weight loss, the potentiodynamic polarization and the electrochemical impedance spectroscopy measurements. In addition, determination of the physico-chemical indexes of the seeds oil of *Ceratonia siliqua L*, showed that it is a long chain unsaturated oil. The results indicated that the inhibition efficiency (% IE) of the FCSL formulation increased with increasing both inhibitor concentrations and immersion time. The inhibition efficiency reached a value of 98.6% at 750 ppm of the FCSL formulation. This formulation may be form a filmed and acts as a barrier, which minimize the contact area between metallic surface and corrosive solution. This leads to decrease the oxidation of the iron substrate. The metallic surface, after inhibitor treatment has been observed using SEM coupled with the EDX analysis. The obtained results indicate that the FCSL acts as an excellent mixed-type inhibitor.

Keywords- Corrosion inhibitor, Green formulation, FCSL, *Ceratonia Siliqua L*, Iron substrate, Rainwater

1. INTRODUCTION

Corrosion is a degradation of metallic materials by chemical interaction with their environment. It is not easy to eliminate and it affects most of the industrial sectors, which this phenomena may cost the billions of dollars each year for preventing and replacement of maintenance [1]. The most efficient method for protection of a lot of metals and alloys is using the corrosion inhibitors against acid attack in various domains of application as a pickling agent, a cleaning agent, an acid descaling and oil well acidizing. The organic molecules are the inhibitors that have the oxygen, sulphur or nitrogen atoms in their structures.

Recently, the plants extracts are used as a safe and non-toxic inhibitor of low carbon steel in acidic solution. Such as M. Zouarhi et al have study, the inhibiting Effect of a Green corrosion Inhibitor Containing *Jatropha Curcas* Seeds Oil for Iron in an Acidic Medium [2]. In addition, H. Hammouch et al used *Opuntia Ficus Indica* seed oil in corrosion inhibition of carbon steel in acid medium [3]. A. M. Abdel-Gaber et al have employed some plant extracts as corrosion inhibitor for steel in acidic media [4]. The use of natural compounds as corrosion inhibitors has been widely reported by several authors, L. Bammou et al [5], A. Bouyanzer et al [6], A. Chetouani et al [7], P. Saxena et al [8], L. Lahhit et al [9], M. Chellouli et al [10]. Also the others authors apply a green inhibitor of corrosion in acidic media [11-16]. In this context, we developed a new non-toxic, and biodegradable corrosion inhibitor.

The electrochemical impedance spectroscopy (EIS) and potentiodynamic polarization techniques have been successfully used for anti-corrosive analysis of the protective coatings and films applied to metals [17].

This work relies on the application of a non-toxic and environmently friendly corrosion inhibitor derived from *Ceratonia siliqua* L as a green corrosion inhibitor of iron substrate in simulated rain water solution.

2. MATERIALS AND METHODS

2.1. Oil extraction process

Before the extraction process, the seeds of *Ceratonia siliqua* L were grounded using a mechanical grinder and then the extracted lipid was obtained by a soxhlet apparatus. The solvent using in this process is the cyclohexane. This later was removed using rotary evaporator apparatus at 313 K. The extracted seeds oil was stored at 269 K.

2.2. Physico-chemical analysis

The physico-chemical characteristics of the *Ceratonia siliqua* L oil such as refractive index, iodine index, density, saponification index, and acid index were determined according to the AFNOR standards respectively [18].

2.3. Electrode, electrolytic solution and corrosion inhibitor

The chemical composition (wt %) of the iron-based coupons is given in the Table 1.

Table 1. Chemical composition of the iron substrate used in this study

Element	Si	Mn	C	P	S	Fe
Wt (%)	0.2	0.519	0.157	0.007	0.009	Balance

The electrolyte composition used in this study was 0.2 g L⁻¹ Na₂SO₄, 0.2 g L⁻¹ NaHCO₃ and 0.2 g L⁻¹ NaCl the solution acidified by addition of the sulfuric acid at pH=3,6.

This solution corresponds nearly to the rain water in urban area, near seaside with an extreme degree of pollution [19].

The corrosion inhibitor used in this work is a new formulation based on *Ceratonia Siliqua* L seed oil noted FCSL. The unit of inhibitor concentration used is ppm. Therefore, we prepared a solution of 1000 ppm by solubilized 1 g of the FCSL formulation in one liter of corrosive solution, then diluted to different concentrations: 750, 500 and 250 ppm.

2.4. Weight loss measurements

The dimension of iron samples used for the weight loss measurement is 2cmx1cmx0.1cm. Prior to all experiments, the exposed area was mechanically abraded by 800, 1200 and 2000 grades emery papers. The specimens were cleaned thoroughly with distilled and we are used the acetone to degreased then dried.

The weight loss W(mg) was calculated after cleaned and decaped with acid, the samples weight was determined before and after 24 h of immersion time at 293 K without and with various concentrations of the FCSL formulation. The gravimetric experiment was performed by using an analytical balance by 0.1 mg of precision.

Weight loss allowed calculation of the mean corrosion rate, for to have a reproducibility results; we repeat the tests three times.

The corrosion rate (v), and the inhibitor's efficiency (IE%) were calculated by the following equations [20]:

$$v = \frac{w}{s \times t} \quad (1)$$

$$E\% = \frac{v_0 - v}{v_0} \quad (2)$$

Where:

W: The weight loss (mg),

S: The specimen's surface (cm²),

t: The immersion time (h)

v: The corrosion rate (mg. cm⁻².h⁻¹)

v₀ and v are the corrosion rate of iron substrate without and with of the FCSL formulation respectively.

The weight loss measurements were used to estimate the average corrosion rate. Therefore, they have only a qualitative method. Indeed, this method presents an imperfection, because it measures only the metal ions in solution without measuring the ions remained in the corrosion products.

In order to determine the main kinetic parameters associated to corrosion process, the electrochemical techniques such as electrochemical impedance spectroscopy (EIS) and Potentiodynamic polarization were used in this study.

2.5. Electrochemical measurements

Before each experiment, we polished the iron electrode with emery papers using different grades from 400 to 2000, then degrease with acetone and rinsed in distilled water before induction into the corrosive solution. The cylindrical rods, into glass tube of appropriate diameter using epoxy resin, offered an active flat disc shaped surface of 1 cm² geometric area to contact with the electrolyte.

Experiment conducted using three conventional electrodes cell in faradic cage to minimize the electrical noise: the iron substrate as a working electrode; rectangular platinum as a counter electrode and the saturated calomel electrode (SCE) was used as a reference electrode. Before polarization and impedance experiments, the working electrode was inserted in the test solution for 30 minutes to attend a steady state open circuit potential (OCP). The electrochemical measurements were performed by using a BioLogic SP-200 electrochemical apparatus interfaced with a computer.

The cathodic and anodic polarization curves were performed using a scan rate equal to 1 mV.s⁻¹. To perform the electrochemical measurement we used the automatic ohmic drop compensation (ZIR). The electrochemical parameters values (I_{cor}, E_{cor} and b_a) were determined using the EC-LAB V 10.36 software.

The calculation of the inhibition efficiency (IE%) from potentiodynamic polarization curves using the following equation:

$$IE \% = \frac{I_{cor0} - I_{cor}}{I_{cor0}} \times 100 \quad (3)$$

Where: I_{cor0} and I_{cor} are the corrosion current density in the presence and in the absence of the inhibitor respectively.

Electrochemical impedance studies were carried out by using signals of 10 mV amplitude in the frequency spectrum range between 100 kHz and 10 mHz at the stable open circuit potential (OCP).

The registration of the EIS spectra started after 30 minutes of immersion time. Then the impedance data were examined by the EC-LAB program [21].

The inhibition efficiency for the EIS spectra at different concentrations of the FCSL formulation was evaluate by using the following equation:

$$IE \% = \frac{R_p - R_{p0}}{R_p} \times 100 \quad (4)$$

Where R_{p0} and R_p are the polarization resistances for the uninhibited and inhibited solutions, respectively [22].

2.6. Surface analysis (SEM)

Scanning Electron Microscopy (SEM) was performe using a LEICA STEREOSCAN 440 electron microscope with EDX elemental analysis.

The electrode was examined after 24 h of immersion time in the presence and in the absence inhibitor; the specimens were washed with distilled water and dried at ambient temperature.

3. RESULTS AND DISCUSSION

3.1. Physicochemical characteristics of *Ceratonia Siliqua L* seeds oil

The physico-chemical characteristics of the seeds oil of *Ceratonia Siliqua L* were reported in table 2.

Table 2. Physico-chemical characteristics of the seed oil of *Ceratonia Siliqua L*

Physico-chemical characteristics	Present study
Density at 293 K	0.9
Refractive index at 293 K	1.3
Saponification index (mg of KOH/g)	178.1
Acid index (mg of KOH/g)	6.3
Iodine index (g/100 g)	150.4

The physico-chemical parameters used in this study give more information about the seeds oil of the *Ceratonia Siliqua L*.

The refractive index increases with the insaturation and the average length of the fatty acid chains, furthermore, the acid index is relatively large. From these results, we can conclude that the seeds have undergone an oxidative alteration during their storage.

3.2. Weight loss measurements

The corrosion rate (v) and the percentage of the inhibition efficiency (% IE) determined by the gravimetric method for different concentrations of FCSL formulation, are presented in table 3.

Table 3. Evolution of the inhibition efficiency and corrosion rate at different concentration of the FCSL

[FCSL] ppm	Blank	250	500	750
Δm (mg)	2.5	0.5	0.4	0.2
v ($\text{mg}\cdot\text{cm}^{-2}\cdot\text{h}^{-1}$)	0.057	0.012	0.009	0.005
IE (%)	---	78.9	84.2	91.2

According to the table 3, the corrosion rate decreases with increasing inhibitor concentration. Indeed the inhibition efficiency reached a value of 91.2% at 750 ppm of the inhibitor. We can conclude that the FCSL formulation has an excellent protection effect against iron corrosion in acidic solution.

3.3. Electrochemical measurement

3.3.1. Open circuit potential

The results of the open-circuit potential variation (OCP) of the iron substrate in acidic solution in presence and absence of FCSL are reported in Fig.1.

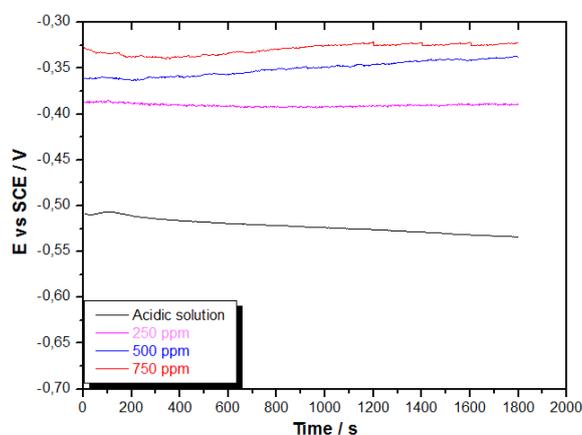


Fig. 1. Variation of the open-circuit potential (OCP) of the iron substrate in acidic solution at various concentration of the FCSL formulation

The results show that in absence of FCSL the potential tends to stabilize at -0,51 V, after 20 min.

The addition of FCSL leads to a shift corrosion potential to positive direction and this increase with increasing of the inhibitor concentration.

This important shift of corrosion potential may indicate an important anodic inhibiting effect of FCSL.

3.3.2. Potentiodynamic curves

The polarization studies of iron were carried out in acid rain solution in both the absence and the presence at various concentration of FCSL formulation.

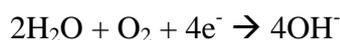
All of these curves were obtained after 30 min immersion time of the electrode in electrolytic solution at corrosion potential (E_{cor}) and after performing the automatic ohmic drop compensation (ZIR).

3.3.3. Cathodic polarization

The Cathodic polarization curves of iron in simulated acid rain solution without and with various inhibition concentrations are reported in Fig. 2a.

In absence of the inhibitor, the corrosion current increase rapidly with the cathodic overvoltage until the potential value of -0.8 V/SCE, for more negative potential values a pseudo plateau appears in 0.4 mA/cm^2 , which can be attributed to the oxygen diffusion process, so the cathodic reaction can be expressed by the following equation:

Therefore, in the cathodic process the important factor must be considerate is mass transport [24].



The adding of the formulation to the corrosive solution is accompanied by both a shift of corrosion potential E_{cor} toward more positive potential, and a decrease of the current density I_{cor} , with the disappearance of the diffusion plateau, obtained in the case of the blank solution, at the concentration of 750 ppm of the FCSL formulation. We observe the formation of the film on the area, which hinders the diffusion of dissolved oxygen towards the electrode surface.

3.3.4. Anodic polarization

The anodic polarization curves of iron in solution-simulated acid rain without and with various inhibition concentrations are reported in Fig. 2b.

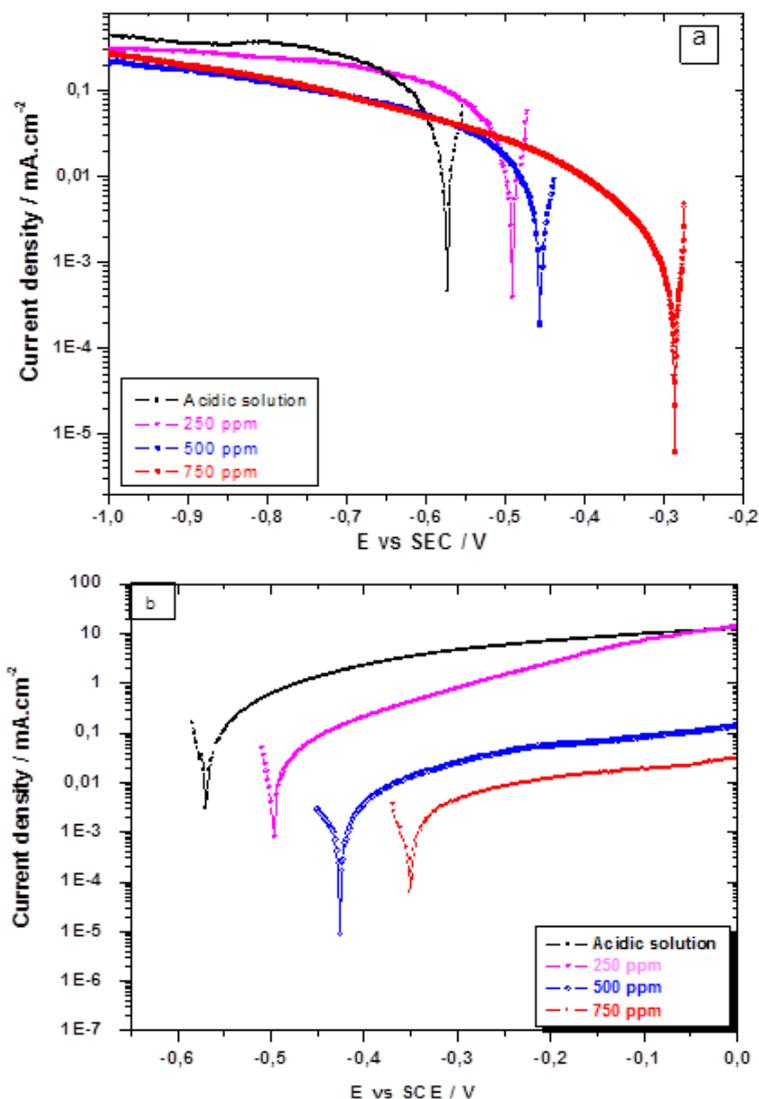


Fig. 2. (a) Potentiodynamic polarization curves of iron in acidic solution at different FCSL concentrations in the cathodic domain (a), after Ohmic drop compensation (ZIR); (b) Potentiodynamic polarization curves of iron in acidic solution at different FCSL concentrations in the anodic domain, after Ohmic drop compensation (ZIR)

This behavior is associated with presence of chloride in solution. The anodic reaction can be expressed by the following equation:



According to the Fig. (2b) in the presence of the FCSL formulation, the current density decreases with increasing of the inhibitor concentration. Furthermore, the value of the corrosion potential displayed to values more positive compared, also the value of the current density I_{cor} reduced from $74.9 \text{ mA}\cdot\text{cm}^{-2}$ in the absence of inhibitor to $1.0 \text{ mA}\cdot\text{cm}^{-2}$ at 750 ppm

of the FCSL formulation. So less than 75 times then we observe the appearance of a current plateau in a wide potential range.

The corresponding current plateau value is in the order of $0.03 \text{ mA}\cdot\text{cm}^{-2}$ at 750 ppm of the FCSL formulation. This may be indicates that the iron surface is protected by the inhibitor; this protection may be attributed to a passivity of iron substrate resulting from the formation of inhibitor film on the iron electrode surface. H. Hammouch and al, M. Chellouli and al observed this inhibition behavior [3-10].

The kinetic parameters of the corrosion of the iron substrate electrode were determined by the EC-lab program. The obtained results are summarized in table 4.

Table 4. Kinetic parameters determined from polarization curves

FSCL (ppm)	E_{cor} (mV/SCE)	I_{cor} ($\mu\text{A}/\text{cm}^2$)	β_a (mA/dec)	IE (%)
Blank	-572	74.9	19.1	----
250	-497	15.3	9.5	79.6
500	-436	1.7	20.6	88.9
750	-330	1.0	22.6	98.6

As shown in Table 4, we note an increasing of the inhibition efficiency with increasing of the FCSL concentrations. This increasing is accompanying by decreasing of the current density I_{cor} . The inhibition efficiency reached a maximum value of 98.6 % at 750 ppm of inhibitor.

From this result, we can conclude that the FCSL formulation is a mixed type inhibitor that acts by decreasing of the current density in both the cathodic and the anodic domain and the corrosion potential become more anodic.

This good inhibiting effect of the FCSL formulation may be related to the adsorption on the electrode surface by the establishment of a barrier film.

3.4. Electrochemical Impedance Spectroscopy

The EIS experiments were carried out to understand the phenomenon interface metal/solution.

The mechanistic information can be provided from the diagram of the impedance spectra [25]. Also the data analysis can be performed by the proper choice of structural models of the interface [27,28], and the obtained results are extensively used for the investigation of the corrosion inhibition processes [29,30]. The EIS measurements were performed as a function of the concentration of the inhibitor and immersion time.

3.4.1. Effect of the concentration of FCSL formulation

The impedance diagrams in Nyquist plot in the absence and the presence of various concentration of the FCSL at 293 K are represented in Figs. 3.

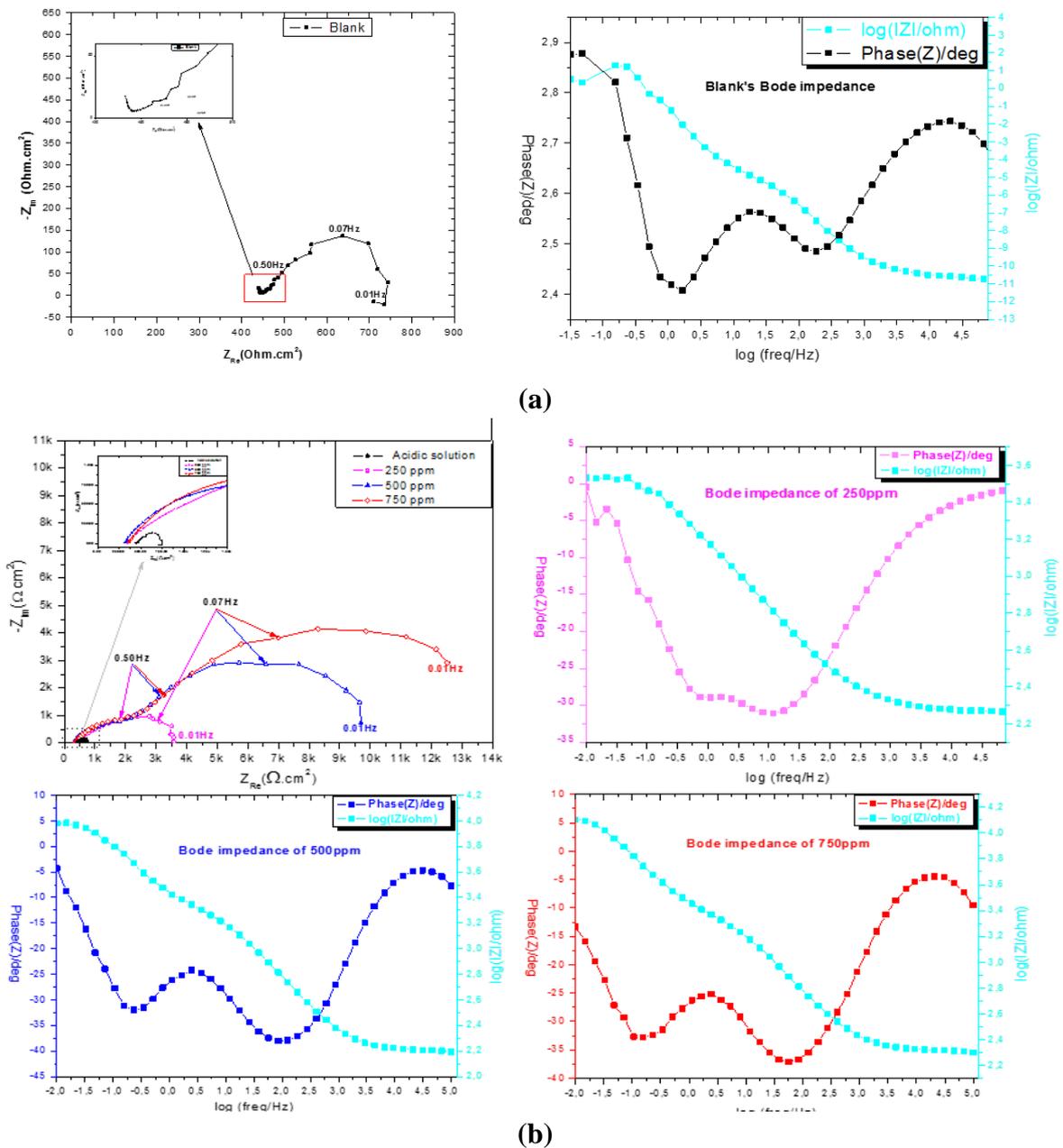


Fig. 3. (a) Nyquist and bode impedance plot of the iron electrode in acidic solution at 293 K; **(b)** Nyquist and bode impedance plots recorded for iron in acidic solution as a function of the FCSL concentrations at 293 K

In the case of the blank solution, as shows in Fig. 3a, we noted the two capacities loops in the high frequencies and the inductive loop at low frequencies [29]. This inductive effect may be due to the desorption of H^+ ions and salts ions present in the solution [30 and 31], or to the

re-dissolution of the passivity surface [32]. In effect, this inductive loop is disappearance with addition of the various inhibitor concentrations (Fig. 3b). The same behavior has been observed by others authors [2,10].

As shown in fig.3b, in the presence of various concentrations of the FCSL formulation, the size of the loops increased with increasing of the concentration of FCSL formulation. We used the polarization resistance to evaluate the inhibition efficiency (% IE).

We noted also, decreasing of the electrolyte resistance may be explicated by the presence of the ionized substances in the formulation.

The electrochemical parameters associated with the EIS diagrams represented in Figs. 3a and 3b are shown in tables 5a and 5b.

Table 5a. Electrochemical parameters associated with the EIS diagram of the blank solution

	R_e ($K\Omega.cm^2$)	R_{HF} ($K\Omega.cm^2$)	C_{HF} ($\mu F/cm^2$)	R_{BF} ($K\Omega.cm^2$)	C_{BF} ($\mu F/cm^2$)	R_p ($K\Omega.cm^2$)
Acidic solution	0.433	0.164	82	0.216	702	0.38

Table 5b. Electrochemical parameters associated with the EIS diagrams presence of various concentrations of FCSL formulation

FCSL concentrations (ppm)	R_e ($K\Omega.cm^2$)	R_f ($K\Omega.cm^2$)	C_f ($\mu F/cm^2$)	R_t ($K\Omega.cm^2$)	C_{dl} ($\mu F/cm^2$)	R_p ($K\Omega.cm^2$)	IE (%)
250	0.335	0.97	6.34	2.31	441	3.28	88.4
500	0.290	2.81	3.65	7.67	198	10.48	96.4
750	0.372	2.84	3.30	11.24	157	14.08	97.3

According to the results reported in tables 5a and 5b, we noted that the values of the transfer resistance R_t and the film resistance R_f increased with increasing of the inhibitor concentration, however the values of the film capacity C_f and capacity of the double layer C_{dl} decreased both. This may explained by the decreased of the surface heterogeneity also, the adsorption of the inhibitor molecules on the area substrate forming a layer between the metal and the corrosives ions [34]. Indeed the inhibition efficiency reached a value of 97.3% at 750 ppm of the FCSL formulation.

The equivalent electric circuits (EEC) shown in Fig. 5 were determined by the EC-LAB program from the EIS data fitting. The use of such electrical equivalent circuit is in agreement with previous results [34,10].

We can be ascribed to the following contributions: the high frequency contribution (C_f, R_f) can be attributed to the dielectric character due to the formation of the film on the iron surface in presence of the inhibitor. The low frequency contribution can be attributed to the double layer capacitance (C_{dl}) at the electrolyte/iron interface at the bottom of the pores coupled with the charge transfer resistance (R_t) [10].

In the case of the blank solution, the equivalent electric circuit given in Fig. 4 shows the presence of the inductive element L in the absence of the FCSL formulation.

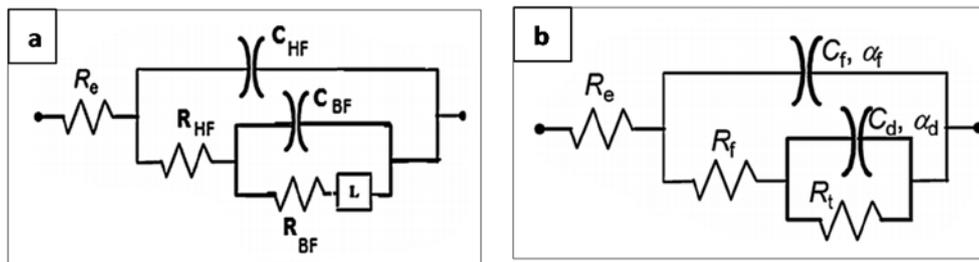


Fig. 4. Scheme of the equivalent electric circuits (EEC) used for iron in acidic solution in the absence (a) and the presence (b) of the FCSL formulation

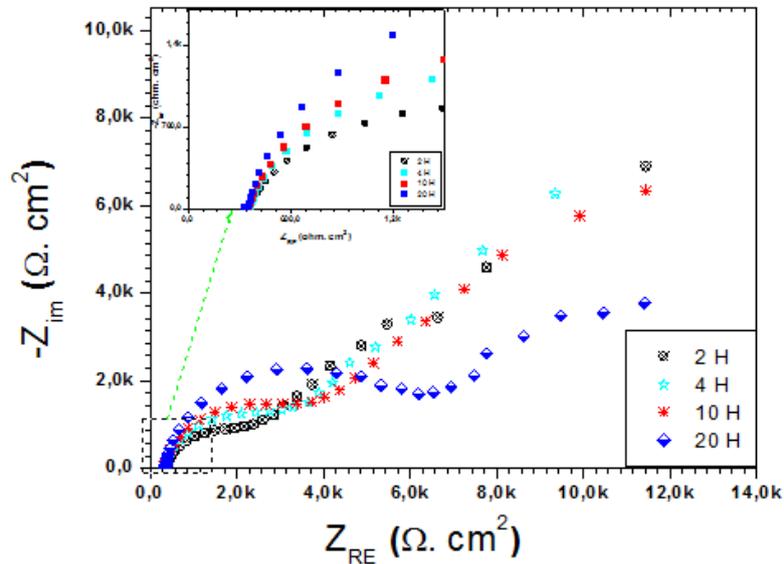


Fig. 5. EIS spectra of the iron electrode in acidic solution at 750 ppm of the FCSL formulation as a function of the immersion time at 293 K

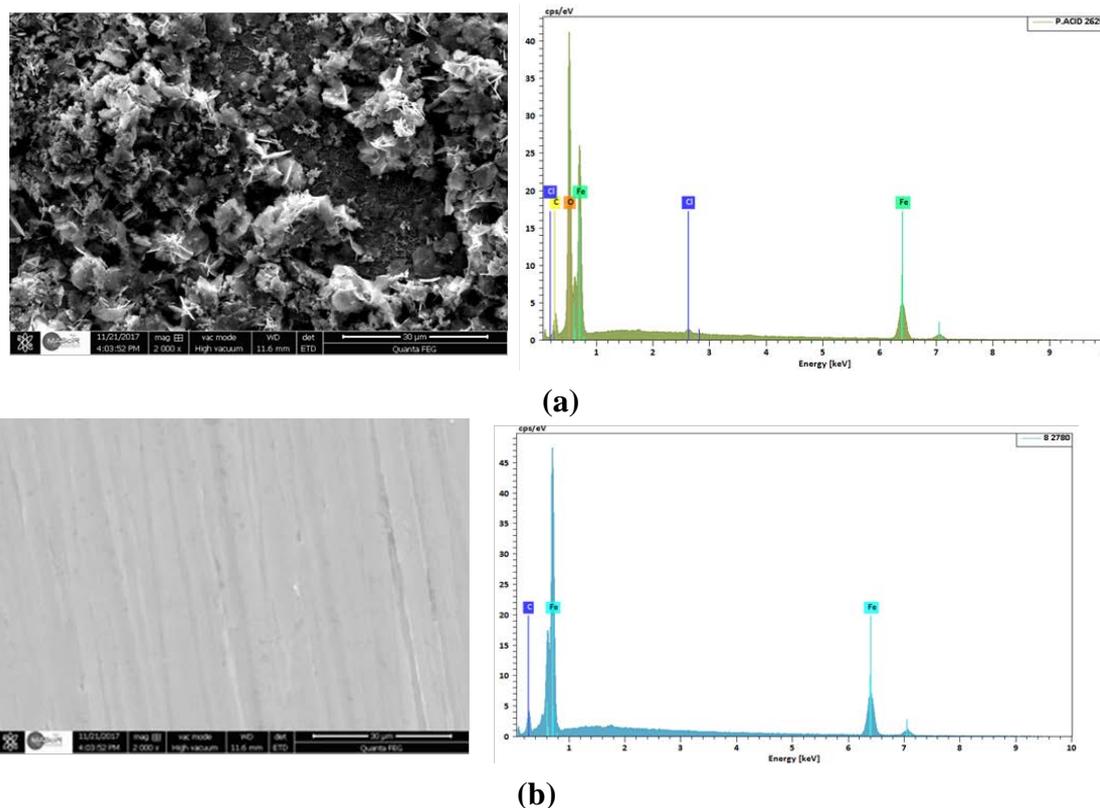


Fig. 6. (a) SEM micrograph and EDS Spectrum of the iron substrate in corrosive solution after 24hour of immersion time in absence of the FCSL formulation; (b) SEM micrograph and EDX Spectrum of the iron substrate in acidic solution after 24 h of immersion time in presence of 750 ppm of the FCSL formulation

3.4.2. Effect of immersion time

The impedance diagrams obtained in solution acid at 750 ppm of the FCSL formulation at the corrosion potential E_{COR} after exposed the iron electrode at different immersion times are shown in Fig. 5.

From the Fig. 5, we noted that the effect of the immersion time on the impedance diagram was characterized by the increase of the size of the two capacities loops reaching a maximum value at 20 h of immersion time. In addition, the capacitive loops maintain the same shape. Although, we observe a significant influence on the size of the impedance diagram with increasing of immersion time.

These results indicate that the FCSL formulation acts as an excellent barrier protecting the iron substrate against the corrosion in acidic medium. To confirm this result, we used the SEM analysis.

3.5. Surface analysis

The aim of the surface analysis by SEM coupled with the EDX used in this study is firstly to check the hypothesis of the formation of the inhibitor on the electrode surface and secondly to verify its protective qualities against iron corrosion.

Figs. 6a and 6b shows the area of the substrate of iron without and with 750 ppm concentration of the FCSL formulation after 24 h immersion time in the acidic solution.

In the absence of the FSCL formulation Fig. 6a, The SEM examination provides that the metallic surface is heavily attacked by the ions corrosive. The EDX spectrum reported in Fig. 6a showed the characteristic peaks of the specimen and a marked presence of the presence of oxygen atoms.

Also in the case of the presence the FSCL formulation Fig. 6b, the addition of the concentration optimal of the FSCL formulation into the corrosion solution, a smooth surface noted this could be explaining the good protection effect of the inhibitor by a formation of the film. As confirmed by the EDX spectrum revealing a very low content of oxygen species.

4. CONCLUSION

In this study the determination of the physico-chemical indexes of the seeds oil of *Ceratonia Siliqua L* showed that it's a long chain unsaturated oil.

The gravimetric test showed that the FSCL formulation has an excellent corrosion inhibiting properties of the iron substrate in the acidic solution. The inhibition efficiency reached a value of 98.6%.

The potentiodynamic curves showed that the inhibiting effect of this compound mark a significant decrease of the cathodic and the anodic current density values especially in the vicinity of corrosion potential.

The inhibition efficiency reached maximum values of 98.6% and 97.3% for the potentiodynamic and the electrochemical impedance spectroscopy measurements respectively at 750 ppm of the FCSL formulation. Also, the size of the diagram of the Nyquist diagram increase with increasing of the immersion time.

The electrochemical measurements showed that the FCSL formulation is an excellent corrosion inhibitor, against iron corrosion in rain acidic solution.

The inhibitor acts as a mixed type inhibitor in both the cathodic and the anodic electrochemical processes.

The good protective effect of the FCSL formulation is attributed to the establishment of the film of inhibitor on the surface of iron substrate. This result was confirmed by SEM analysis coupled with the EDX.

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