Full Paper

The Study of Corrosion Inhibition Mechanism of one of the Salvia Officinalis Extract on Carbon Steel in H_2S and HCl Solutions

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Abstract- Corrosion inhibition of carbon steel in solutions containing H_2S and HCl by using the Salvia officinalis extract was studied by the Tafel polarization and electrochemical impedance spectroscopy (EIS) methods. Tafel polarization and EIS results revealed that the protection efficiency increases with increasing inhibitor concentration to attain 93% at 7.5 g/l inhibitor. Also, the Tafel polarization plots showed that current inhibitor acts as a mixed type inhibitor. It was found that inhibition occurs due to the physical absorption of inhibitor on the metal surface that follows from longmuir isotherm.

Keywords- Corrosion inhibition, Carbon steel, Salvia officinalis extract, Electrochemical impedance spectroscopy, Tafel polarization

1. INTRODUCTION

Since carbon steel is widely used in chemical reactors, oil and gas transport pipelines, heat exchanger and storage tanks, corrosion of this alloys has been one of the major concerns [1,2]. Generally, corrosion of metals and alloys in H_2S and HCl solutions is very important and serious in petroleum industries [3]. Indeed, acetic acid, chlorides and hydrogen sulfide (H_2S) are the most corrosive reagent (NACE solution TM 0177) in primary distillation factories that can create a corrosive solution with crude oil [4,5]. H_2S corrosion has been considered by researchers for decades [6]. Up to now, several studies have been performed

related to the corrosion of steel in H_2S and HCl solutions [7]. For example, Veloz and González [8] have evaluated the corrosion behavior of a steel in acetic acid solutions with chlorides and H_2S. In chemical and petroleum industries, many ways (such as adding inhibitor) are used to prevent or reduce of corrosion [9–11]. Many corrosion inhibitors that are used in acidic media are organic compounds that have electronegative atoms such as oxygen, nitrogen, sulfur and unsaturated bonds and aromatic rings [12,13]. But most of these inhibitors are toxic, non-renewable and expensive that can be a serious threat to environmental and human health [9,10]. Among these compounds, plant extracts for decades have been favorite subject of many researchers [14].

Different parts of a plant such as root, seed, stalk, leaves and flower can be used as a corrosion inhibitor [14,15]. Some basic ingredients from plants that have been reported as a good corrosion inhibitor include sugars, steroids, aloin, tannic acid, flavonoids, terpenes and etc. The presence of polycyclic compounds accelerates film formation and reducing corrosion on steel surface [15–20].

Salvia officinalis (Fig. 1) is a plant that belongs to the family Lamiaceae and in the past has been of particular interest in the field of medicine. It has been used in pharmaceutical, perfume and cosmetic products and food industry. Common sage is a perennial herbaceous plant, root right. It has many branches shoot straight, and a height of 50 to 80 cm with dark green young stems. Salvia officinalis grows in dry and rocky different areas of Asia, North Africa, Mexico and Iran [21].
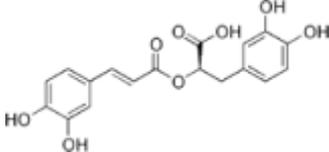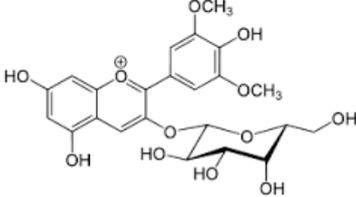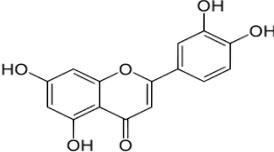

Fig. 1. Picture of Salvia officinalis

Some of the Salvia officinalis extract compound are monoterpene such as Limonene, Terpinolen, Camphene and diterpene such as Carnosol, Tanshinone, Ursolic acid, and flavonoid such as Apigenin and Sesquiterpenoids and other Salvia officinalis's compounds [22–24]. Most components of the present extract are phenolic compounds, contain

heteroatom oxygen and aromatic rings. The structure of some of the most important compounds has coming in Table 1. It seems that Salvia officinalis, due to these compounds, can easily absorbed on the steel surface and acts as a good corrosion inhibitor.

Table 1. Some chemical structures of Salvia officinalis components

Name	Compounds
Rosmarinus *acid*	
Anthocyanin	
Luteolin	

Based on a literature survey, no previous work has been reported on the corrosion inhibition of Salvia officinalis extract for mild steel in H_2S and HCl solutions. The present research, to the best of our knowledge, is the first of its kind that focuses on the effect of Salvia officinalis extract on the corrosion behavior for mild steel in H_2S and HCl solutions. For this purpose, the EIS and tafel polarization measurements were carried out. Also, this study focuses on the mechanism of inhibition based on adsorption isotherms and thermodynamic parameters.

2. EXPERIMANTAL PROCEDURE

2.1. Preparation of Solution and Extract of Salvia Officinalis

Firstly, base solution (0.25 M Na_2SO_4+0.5 M H_2SO_4+0.5 M Na_2S) was prepared in distilled water. For this purpose, analytical reagent grade sodium sulfate, sulfuric acid, sodium sulfide and hydrochloric acid were used. Then, 198.99 mg/l H_2S must be created in the base solution. For the preparation of the 198.99 mg/l H_2S solution containing 0.01 wt%

HCl, 2.543 ml of 0.5 M Na_2S solution and the same volume of 0.5 M H_2SO_4 solution were added into 100 ml Na_2SO_4 solution, and then 0.025 ml of 37 wt% HCl was added to the solution.

The plant of Salvia officinalis was dried in oven at 60 °C and grinded to powdery. The powder (20 g) was refluxed in double distilled water (400 ml) for 10800 s. Then, the obtained solution was filtered to remove any contamination. The extract of the Salvia officinalis was prepared by evaporating the filtrate on water bath and drying the paste residues in desiccator.

2.2. Electrochemical Measurements

Chemical composition of SAE 1018 carbon steel used in this experiment is shown in Table 2. All samples were polished to 1200 grit and cleaned by distilled water prior to each test. The electrochemical experiments were performed in a conventional three-electrode cell using µAutolab Type III/FRA2 system controlled by a personal computer. A platinum wire, an Ag/AgCl electrode (in saturated KCl solution), and SAE 1018 carbon steel samples were used as the counter electrode, reference electrode, and working electrodes, respectively. All experiments were performed at room temperature in test solution prepared from analytical grade chemicals and distilled water. Prior to each electrochemical test, the specimens were exposed to test solution for 900 s until the steady corrosion potentials were obtained. The electrochemical measurements were performed in the following sequence: (a) Tafel polarization tests were carried out starting from –0.25 V (*vs*. E_{corr}) and ending at 0.25 V (*vs*. E_{corr}), and (b) EIS tests were recorded over the frequency range 100 kHz to 10 mHz and with a 10 mV amplitude. NOVA software was used for modeling the EIS data.

Table 2. Chemical composition of SAE 1080 carbon steel

	C	Mn	B	Cu	Pb	Fe
SAE 1080 steel / wt%	0.15	0.60	0.3	0.2	0.1	Bal

3. RESULTS AND DISCUSSION

3.1. Tafel Polarization Measurement

Effect of Salvia officinalis extract concentration on the corrosion behavior of SAE 1018 carbon steel samples in test solutions (containing 198.99 mg/l H_2S and 0.01 wt% HCl) has been evaluated by Tafel measurements and the recorded Tafel plots are shown in Fig. 2. From these curves, the electrochemical parameters as obtained are shown in Table 3. These include corrosion potential (E_{corr}), corrosion current density (i_{corr}) and cathodic Tafel slope

(B_c). Corrosion current density determined by extrapolation of the cathodic branch to the corrosion potential. Moreover, the inhibition efficiency ($E_p\%$) can be measured using Eq. (1) [25,26]:

$$E_P\% = \frac{i_{corr}^0 - i_{corr}}{i_{corr}^0} \times 100 \qquad (1)$$

Where i_{corr} is the value of corrosion current density in the presence of the inhibitor and i_{corr}^0 is the value of corrosion current density in the absence of the inhibitor.
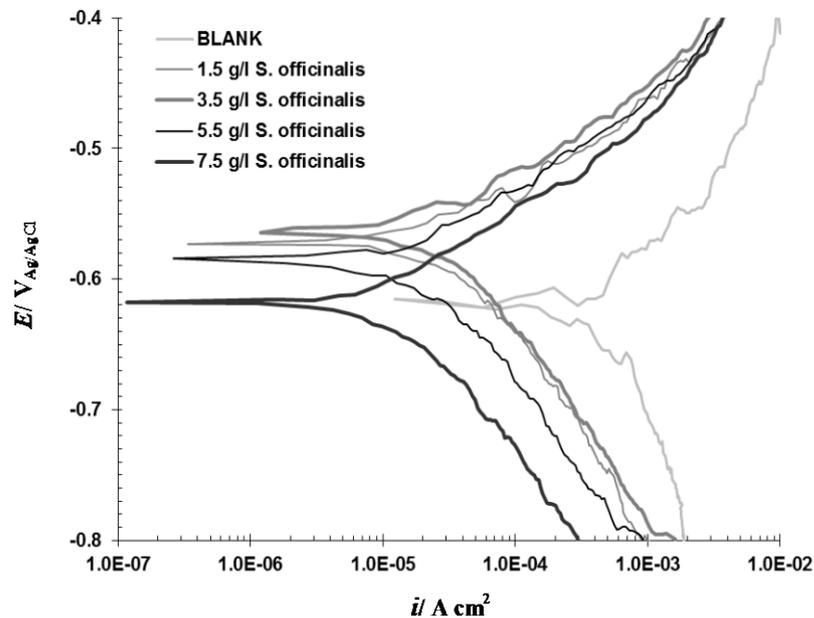

Fig. 2. Tafel polarization curves of SAE 1018 in the absence and presence of Salvia officinalis extract in H_2S and HCl solutions (scan rate=1 mV/s)

The results show that corrosion current density decreased with increasing inhibitor concentration that the maximum decrease in the concentration of 7.5 g/l was obtained. Both anodic and cathodic current density is decreased in the presence of inhibitor, while cathodic branch more than anodic is affected by increasing inhibitor concentration. In another way if an inhibitor changes the E_{corr} more than 85 mV, then can be classified to anodic or cathodic type [26,27].

In this study, corrosion potential shifting is about 59 mV after adding Salvia officinalis, suggesting that this inhibitor acted as a mixed-type inhibitor. It is seen that the corrosion potentials shift toward noble values on the introduction of the extracts into the corrosive solution in comparison to the blank solution.

Table 3. Tafel polarization data for SAE 1018 carbon steel corrosion in the absence and presence of different concentrations of Salvia officinalis in H_2S and HCl solutions

Concentration (g/l)	E_{corr} ($V_{Ag/AgCl}$)	i_{corr} ($\mu A/cm^2$)	β_c (mV/decade)	Θ
-	-0.623	135	-134.1	-
1.5	-0.572	26.32	-70.94	0.81
3.5	-0.564	20.95	-69.01	0.84
5.5	-0.584	10.05	-68.68	0.92
7.5	-0.617	9.03	-67.86	0.93

Also, the displacement of the potentials toward a positive direction was seen to be dependent on the extracts' concentration that is, shifting more toward more positive values with increasing concentration. This could be correlated to the formation of protective film on the surface of the SAE 1018 carbon steel samples as a result of adsorption of the components of the extracts on the surface.

3.2. EIS Measurement

The effect Salvia officinalis extract concentration on the impedance behavior of SAE 1018 carbon steel in solutions containing 198.99 mg/l H_2S and 0.01 wt% HCl is studied. Fig. 3 shows the Nyquist and bode plots that recorded for the corrosion inhibition of SAE 1018 carbon steel with different concentrations of Salvia officinalis extract in H_2S and HCl solutions. The Nyquist plots were obtained with depressed semicircle in the absence and presence of inhibitor. The results show that the diameter of the semicircles increased with increasing Salvia officinalis extract concentration. Indeed, the depression in the semicircles can be correlated to the surface roughness and adsorption of the inhibitor on the alloy surface [28].

Fig. 3b and c show the bode and bode-phase plots, respectively, for the corrosion of SAE 1018 carbon steel in the presence of different concentrations of Salvia officinalis extract. It is seen that the phase angle increases by adding Salvia officinalis extract up to an optimal level. Generally in the Bode plot, the difference between the low frequency and high frequency limit is equal to polarization resistance (R_p). It is seen that the difference between the high frequency and low frequency for the uninhibited and inhibited systems increases with increase in the inhibitor concentration of Salvia officinalis extract up to the critical concentration (Fig. 3b). The increase in phase angle with increase in the Salvia officinalis extract concentration (Fig. 3c) is correlated to the decrease in the capacitive behavior at the metal surface due to decreased metal dissolution rate. This can be related to adsorption of, the main constituents of Salvia officinalis extract that having number of hydroxyl group,

heteroatom oxygen and large number of aromatic rings. Also, the good performance of this inhibitor can be due to large size of constituent's molecule [24].
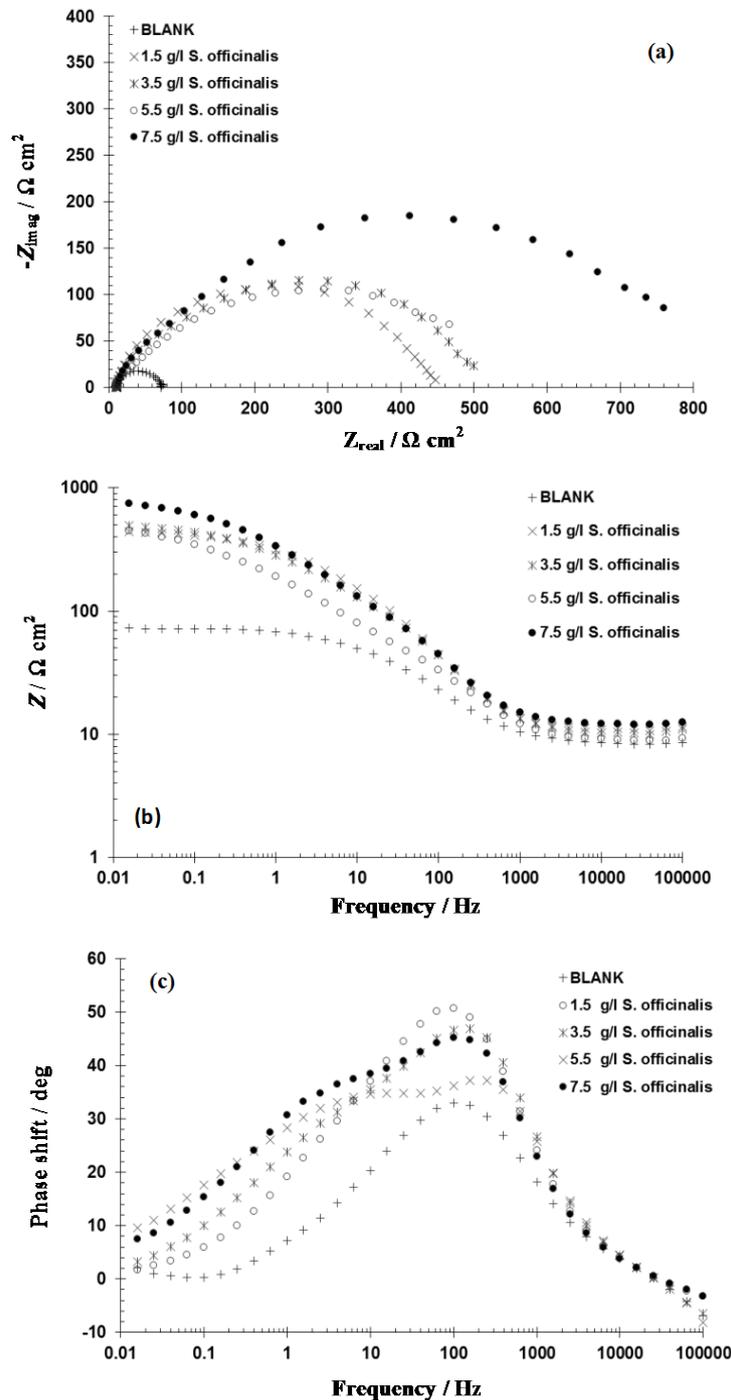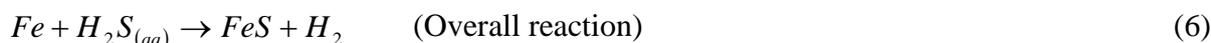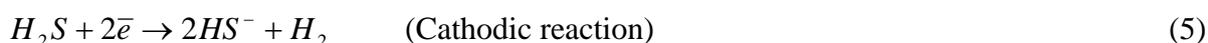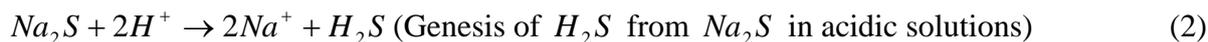

Fig. 3. (a) Nyquist, (b) Bode, and (c) Bode-phase plot of SAE 1018 in the absence and presence of different concentrations of Salvia officinalis extract in H2S and HCl solutions

The corrosion reactions between surface and solution when steel is exposed to H2S and HCl solution are as follows [29–39]:

$$HCl \rightarrow H^+ + Cl^- \tag{1}$$

$Na_2S + 2H^+ \rightarrow 2Na^+ + H_2S$ (Genesis of H_2S from Na_2S in acidic solutions) $\qquad$ (2)

$Fe \rightarrow Fe^{2+} + 2\bar{e}$ $\qquad$ (Anodic reaction) $\qquad$ (3)

$2H^+ + 2\bar{e} \rightarrow H_2$ $\qquad$ (Cathodic reaction) $\qquad$ (4)

$H_2S + 2\bar{e} \rightarrow 2HS^- + H_2$ $\qquad$ (Cathodic reaction) $\qquad$ (5)

$Fe + H_2S_{(aq)} \rightarrow FeS + H_2$ $\qquad$ (Overall reaction) $\qquad$ (6)

To analyses the impedance data, the equivalent circuits that shown in Fig. 4 are used. The circuit shown in Fig. 4a is used to fit the EIS plot in the absence of inhibitor in solutions containing 198.99 mg/l H₂S and 0.01wt% HCl. In this equivalent model, R_s is the solution resistance, R_{ct} is the charge transfer resistance, and *CPE* shows the constant phase element. Generally, the constant phase element is used instead of the double-layer capacitance (C_{dl}) to justify the heterogeneity in the system [40]. Moreover, the impedance data in the presence of Salvia officinalis extract simulated by the equivalent circuit that shown in Fig. 4b [41]. In this circuit, R_s is the solution resistance, R_{ct} is the charge transfer resistance, CPE_{dl} is the constant phase element of the electric double layer, CPE_f is the constant phase element of the surface film and R_f is the surface film resistance. Both R_{ct} and R_f increased with increasing in Salvia officinalis extract concentration at ambient temperature. The impedance parameters obtained are shown in Table 4. It must be mentioned that Eq. (2) was used to obtain capacitances, CPE_{dl} and CPE_f [42]:

$$C = \left(Y_0 R_{ct}^{(1-n)}\right)^{1/n} \qquad (2)$$

Where Y_0 is admittance and n is a measure of heterogeneity of surface. The value of n can change between 0 and 1. If the value of n=1, the *CPE* acts like an ideal capacitor [40,42].

Table 4. EIS parameters for SAE 1018 steel corrosion in the absence and presence of Salvia officinalis extract in H₂S and HCl solutions

Inhibitor concentration (g/l)	R_s (Ωcm^2)	R_f (Ωcm^2)	R_{ct} (Ωcm^2)	R_p (Ωcm^2)	C_f ($\mu F cm^2$)	C_{dl} ($\mu F cm^2$)
0	8.11	-	64.3	64.3	-	147.2
1.5	10.7	7	426	433	17.6	127.8
3.5	10.4	12	510	522	17.1	122.4
5.5	9.5	16	524	540	16.7	120.8
7.5	12.2	63.9	752	815.9	16.6	115.3

The results show that n in the CPE_{dl} and in the CPE_f was nearly equal to 0.6 and 0.9 respectively, and the behavior of *CPE* reflected non-ideal capacitance at this instance [43]. It

is obvious from results that the value of C_{dl} decreases from 147.2 to 115.3 $\mu F cm^{-2}$ with adding and increasing Salvia officinalis extract concentration. This is related to the increase in the electrical double layer at the metal/ solution interface. This process shows that the mechanism of inhibition is adsorption of inhibitor on the metal/solution interface. Replacement of water molecules by inhibitor molecules on the surface can be another reason for decreasing in the C_{dl} values [44–46].
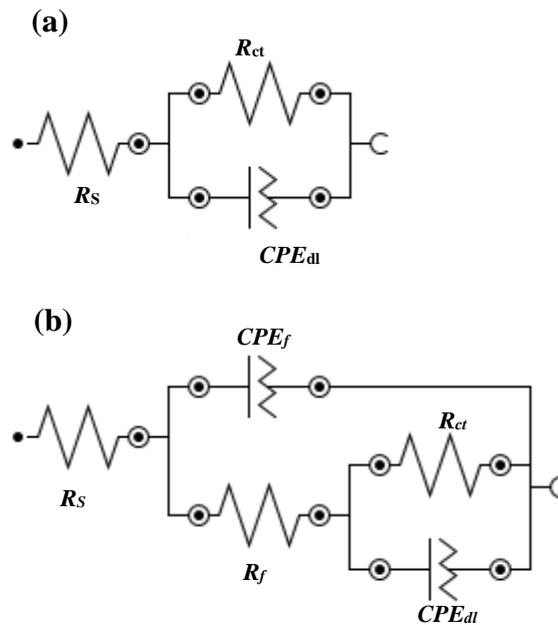

Fig. 4. The equivalent circuits used to model the experimental EIS data of SAE 1018 in H_2S and HCl solutions in the (a) absence of Salvia officinalis, and (b) in the presence of Salvia officinalis

The polarization resistance is calculated by the summation of R_{ct} and R_f at different concentrations of inhibitor. Protection efficiency obtained from Eq. (3) [47,48]:

$$IE\% = \frac{R_P - R_P^0}{R_P} \times 100 \tag{3}$$

Where R_p and R_p^0 indicate the polarization resistance in the presence and absence of Salvia officinalis extract, respectively. The values of R_p increased with increasing inhibitor concentration. Protection efficiency which extracted from polarization and one that obtained from impedance data were compared with each other in Fig. 5.
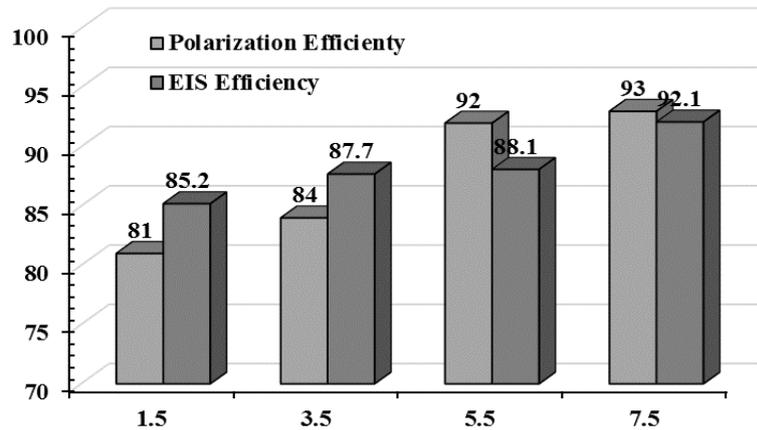
Fig. 5. The chart of comparison between efficiency obtained from polarization data and impedance parameters

3.3. Adsorption Isotherm

Adsorption isotherm is one of the ways to know the mechanism of interaction between inhibitor molecules and metal surface [49]. Langmuir isotherm was selected as the best isotherm and characterized by Eq. (4) [50–55]:

$$\frac{C_{inh}}{\theta} = \frac{1}{K_{ads}} + C_{inh} \tag{4}$$

Where C_{inh} shows the extract concentration, θ represents the degree of surface coverage and K_{ads} illustrate the equilibrium constant of adsorption–desorption process. Langmuir isotherm plot is seen in Fig. 6. Generally this isotherm postulates monolayer adsorption of inhibitor on the surface. In practice, plant extracts are comprised of many organic compounds that can adsorbed differently on the surface. Generally, standard Gibbs free energy (ΔG^0_{ads}) of the adsorption process obtained from Eq. (5). It must be mentioned that K_{ads} is determined from the intercept of the Langmuir isotherm plot [56–58].

$$\Delta G^0_{ads} = -RT \ln\left(55.5 \times K_{ads}\right) \tag{5}$$

Where R shows the molar gas constant, T represents the absolute temperature and 55.5 is the concentration of water expressed in mol/dm^3. If the magnitude of ΔG°_{ads} approximately – 20 kJ/mol or less negative suggesting physical bonding between inhibitor and the charged metal surface (i.e., physisorption). ΔG^0_{ads} around –40 kJ/mol or more negative is an indication of charge sharing or charge transferring from an organic specie to the metal surface to form a coordinate type of metallic bond (i.e., chemisorption) [54]. The value of ΔG^0_{ads} in this study obtained about –14.156 kJ/mol which shows physical absorption of Salvia officinalis extract on the steel surface.
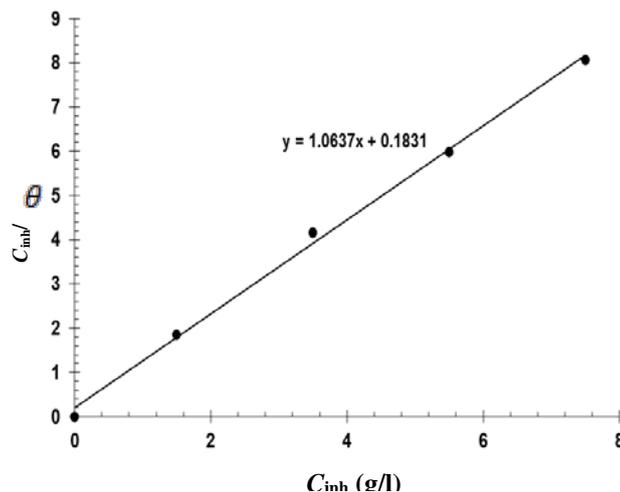
Fig. 6. Langmuir adsorption isotherm for SAE 1018 in the absence and presence of Salvia officinalis extract in H2S and HCl solutions

4. CONCLUSIONS

In this study, corrosion inhibition of SAE 1018 carbon steel in solutions containing H2S and HCl by using the Salvia officinalis extract was studied by the Tafel polarization and EIS methods. The results showed that Salvia officinalis extracts has a significant effect on reducing the rate of corrosion of SAE 1018 carbon steel in H2S and HCl solutions. Also, the results revealed that Salvia officinalis extracts acts as a mixed type inhibitor. Absorption of this inhibitor on the SAE 1018 carbon steel surface follows from Langmuir isotherm. Value of ΔG^0_{ads} indicates that adsorption of present inhibitor on the SAE 1018 carbon steel surface is physically.

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