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# Effect of *Aaronsohnia Pubescens* Extracts to Prevent Against the Corrosion of Mild Steel in 1.0 M HCl

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**Abstract**- The corrosion inhibition and adsorption (CIA) performance of the aqueous extracts of *Aaronsohnia pubescens* aerial parts (Odorized aqueous extract (OE) and Deodorized aqueous extract (DE)) on the corrosion mild steel (MS) in 1M hydrochloric acid were evaluated. It is based on the weight loss (WL) analysis, kinetic and thermodynamic parameters, and electrochemical methods both stationary (Potentiodynamic polarization (PDP)) and transient (Electrochemical impedance spectroscopy (EIS)). The inhibition efficiency of inhibitors increases for the concentration of 1.5 g/L; reaching a high value of 93.11 and 87.88 % in 1 M HCl solution at 308 K for OE and DE, respectively. The thermodynamic kinetic parameters showed that the adsorption of OE and DE on the MS surface follows the Langmuir adsorption isotherm. Furthermore, PDP measurements exhibited that the studied of each inhibitor performs as a mixed-type inhibitor. OE shows itself to be the best aqueous extract of *Aaronsohnia pubescens* aerial parts to prevent against the corrosion of mild steel.

Keywords- Aaronsohnia pubescens; Aqueous extracts; Mild Steel; Corrosion inhibition

## **1. INTRODUCTION**

MS is one of the major building materials that are widely used in industry [1, 2]. In acidic medium, the aggressiveness corrosive compounds attacked this concerned metal. To limit its attack, many organic compounds have proven to be among the most effective inhibitors against corrosion to protect several metals, especially for MS in acidic medium. Therefore, tremendous studies in this field reveal that the most inhibitor deed by adsorption on the electrode surface, and the adsorption behavior of organic compounds does not depend only on the nature of the electrode surface and electrochemical potentials at the metal/solution interface. Besides depends to the structure and electronic properties of inhibitor, which sure influences on the adsorption metal/inhibitor [3].

Almost all inhibitors of corrosion are synthetic organic compounds [4–6], but these compounds are often considered as highly toxic species. Recently, using natural inhibitors has become important because of their environmentally friendly nature [7, 8]. In our laboratory, a great deal of research has carried out to study inhibitive effect several plant extract on the corrosion of steel in 1 M HCl [9–14]. In this work, our focus chiefly put on the aqueous extracts of *Aaronsohnia pubescens*.

*Aaronsohnia pubescens* (Desf.) K. Bremer & Humphries is botanically a synonym of *Matricaria pubescens* (Desf.) Schultz (Asteraceae). It is a pleasant-scented annual plant endemic to North Africa [15] and the Canary Islands [16]. It is primarily found in non-saline wadis on preferably sandy-loamy soils. Parts of the plant are used in local folk medicine: In Morocco, this species, known locally as "*Taraght*", decoctions of the aerial flowering parts are used as mouthwash against toothache [17]. In both Algeria and Morocco, infusions of the aerial parts are used against gastric ulcers and flatulence, respiratory disorders and rheumatic diseases [18]. The main objective of the current research is to study the preventive impact of OE and DE on corrosion of MS in 1M HCl solution using WL measurement, PDP, and EIS.

#### 2. EXPERIMENTAL

#### 2.1. Inhibitors

The experimental phase followed in the current study such as preparations of the aqueous extract (OE and DE) have been mentioned according to a formerly described experimental procedure in our work [19].

## 2.2. Preparation of MS segments, corrosive solutions and inhibitors

Corrosion tests were conducted on the MS segments with dimension of  $2 \text{ cm} \times 2 \text{ cm} \times 0.05$  cm; the MS metal presents the subsequent composition with mass percentage (%): P 0.09, Si 0.38, Al 0.01, Mn 0.05, C 0.21, S 0.05 and Fe 99.21. Before each corrosion test, the MS segments were mechanically ground with sequential grades of emery papers (400, 600 and

1200 grit), and ensuing rinsed and degreased with distilled water and acetone, then dried and weighed. The corrosive acid solution was a molar hydrochloric acid (1 M of HCl), was prepared from concentrated commercial acid of HCl (37%) by dilution with distilled water.

## 2.3. Corrosion monitoring techniques

#### 2.3.1 Weight loss (WL) measurements

The prepared MS samples were weighed; then they were immersed in 1M HCl with and without different concentrations from 0.25 to 1.5 g.L<sup>-1</sup> of inhibitors under different temperatures of 308 to 343 K for an exposure time of six hours [20]. Inhibitory efficiency (IE %) was determined basing on the MS corrosion rates W (mg.cm<sup>-2</sup>.h<sup>-1</sup>) according to the subsequent relationship [21]: (Eqs. 1,2):

$$W = \frac{m_f - m_i}{S.t}$$
(Eq. 1)

where  $m_i$  and  $m_f$  (mg) is the MS sample weight before and after immersion in the tested solution. S is the area of the MS sample (cm<sup>2</sup>) and t is the exposure time (h).

$$IE\% = \frac{W \cdot W'}{W} \times 100$$
 (Eq. 2)

With W and W' are the MS corrosion rates in both uninhibited and inhibited solution, respectively. The extent of the surface coverage ( $\theta$ ) is defined as follows (Eq. 3):

$$\theta = \frac{\text{IE\%}}{100}$$
(Eq. 3)

#### 2.3.2. Electrochemical (PDP and EIS) measurements

Before conducting each electrochemical corrosion test, the sample was immersed in the test solution during 30 min at the open circuit potential (EoCP) to find a steady state. Both PDP and EIS tests were performed from utilizing an assembly of electrochemical cell containing three electrodes: a disc cut form of MS as working electrode (1cm<sup>2</sup>), a saturated calomel electrode as a reference electrode and a platinum electrode as counter electrode. All electrochemical measurements were carried out using a potentiostat/galvanostat (Model 263A) and the electrochemical parameter values were determined using the Volta Master 4 software. We note that all potentials measurement in this study were obtained with respect to the potential value of the reference electrode.

The PDP tests were conducted in 1M HCl and in different concentrations of inhibitors (0.25 to  $1.5 \text{ g.L}^{-1}$ ) in the temperature range of 308 to 343 K, simultaneously. Furthermore, the cathodic and anodic curves were plotted by varying the electrode potential between -800 to +800 mV versus reference electrode at open circuit potential with a scan rate of 0.5 mV/sec.

Inhibitory efficiency  $E_{PDP}$  (%) was calculated based on the current corrosion I of MS at the above conditions according to the ensuing relationship (Eq. 4):

$$E_{PDP} (\%) = \left(\frac{I_{corr} - I_{corr,inh}}{I_{corr}}\right) \times 100$$
 (Eq. 4)

Where  $I_{corr}$  and  $I_{corr,inh}$  are the corrosion current densities in the absence and the presence of inhibitor, respectively. Total charge transfer resistances  $R_t$  (without inhibitor) and  $R'_t$  (with inhibitor) were calculated from Nyquist plots by measuring the impedance difference at low and high frequencies. We maintained the above experiment conditions, and then we conducted the EIS tests in the frequency range of 100 KHz to 10 mHz at open circuit potential with signal amplitude of 10 mV. The inhibitory efficiency  $E_{EIS}$  (%) was calculated according to (Eq. 5).

$$E_{EIS}(\%) = \left(\frac{R'_t - R_t}{R'_t}\right) \times 100$$
 (Eq. 5)

The double layer capacitance (C<sub>dl</sub>) and the frequency at which the imaginary component of the impedance is maximal ( $-Z_{max}$ ) are found determined by Eq. (6):

$$C_{dl} = \frac{1}{2.\pi.f_{\text{max}}.R_t}$$
 (Eq. 6) where  $\omega = 2 \pi$ .  $f_{\text{max}}$ 

 $f_{max}$  is the frequency for which the imaginary part of the impedance is maximum

## 3. RESULTS AND DISCUSSION

## 3.1. Effect of both concentration and temperature

This part aimed to study the influence of OE and DE concentration and temperature media on the progress of corrosion rate of MS and the inhibition efficiency associated to the OE and DE in corrosive media (1M HCl) at range temperature of 308 to 343K without and with different concentrations of inhibitors. The values of percentage inhibition efficiency  $E_w$  (%) and the corrosion rate (W) obtained are summed up in Table 1.

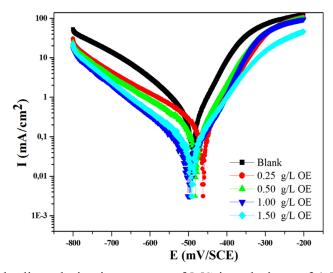
From Table 1, it has appeared that the inhibitor has an inhibiting behavior to limit corrosion on MS surface. Furthermore, we observed that the corrosion rate of MS decreases when the concentration of inhibitor increases and consequently the inhibition efficiency of inhibitors increases for the concentration of 1.5 g/L; reaching a high value of of 93.11 and 87.88 % in 1 M HCl solution at 308 K for OE and DE, respectively. This is due probably to the increase of the surface coverage  $\theta$  for the studied adsorption; thus, it could be ensured the apparition of the defensive deposit, which limits the dissolution of the MS. In other words, we observed that the inhibition efficiency of OE slightly increased when the temperature media increases, which results probably to the chemisorption behavior of OE onto MS surface. The distinction between OE and DE inhibitory effectiveness could be explained by the nature of the molecules of these inhibitors.

	C (g/L)	308 K		313 K		323 K		333 K		343 K	
		W	Ew								
		(mg/cm <sup>2</sup> .h)	(%)								
	1M HCl	0.942		1.751		2.836		3.641		6.301	
0.5	0.25	0.186	80.25	0.344	80.37	0.532	81.24	0.682	81.28	1.115	82.30
OE	0.50	0.116	87.66	0.214	87.78	0.320	88.70	0.409	88.78	0.638	89.88
	1.00	0.108	88.58	0.197	88.74	0.315	88.88	0.404	88.90	0.633	89.96
	1.50	0.065	93.11	0.119	93.22	0.189	93.33	0.241	93.39	0.412	93.46
	0.25	0.224	76.21	0.470	73.15	0.824	70.95	1.064	70.77	1.872	70.29
DE	0.50	0.193	79.50	0.378	78.44	0.622	78.08	0.822	77.42	1.426	77.37
DE	1.00	0.152	83.89	0.309	82.38	0.507	82.12	0.667	81.68	1.170	81.43
	1.50	0.114	87.88	0.276	84.23	0.473	83.33	0.621	82.95	1.230	80.48

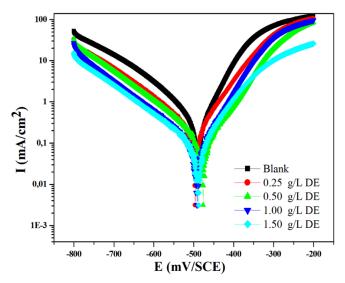
**Table 1.** Weight loss results for MS in 1 M HCl with and without different concentrations of aqueous extracts (OE and DE) at different temperatures

**Table 2.** Electrochemical parameters for the MS in 1 M HCl with and without various concentrations of aqueous extracts from *Aaronsohnia pubescens* aerial parts (OE and DE) at 308 K

	C (g/L)	- E <sub>corr</sub> (mV/SCE)	I <sub>corr</sub> (mA cm <sup>-2</sup> )	-βc (mV dec <sup>-1</sup> )	βa (mV dec <sup>-1</sup> )	EI <sub>PDP</sub> %
1MHCl		490.0	0.5779	147.4	74.8	
	0.25	462.9	0.1145	110.2	60.8	80.18
05	0.50	460.1	0.0857	100.1	67.1	85.17
OE	1.00	445.8	0.0701	109.2	62.9	87.86
	1.50	456.0	0.0517	100.7	65.0	91.02
	0.25	497.8	0.1651	110.6	74.3	71.43
DE	0.50	479.8	0.0997	104.8	73.5	82.74
DE	1.00	477.3	0.0897	110.7	72.1	84.47
	1.50	480.0	0.0862	143.5	74.4	85.05



**Fig. 1.** Anodic and cathodic polarization curves of MS in solutions of 1 M HCl without and with different concentrations of OE



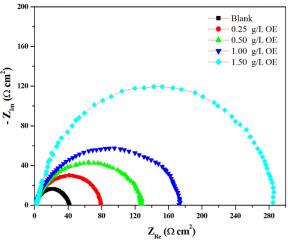
**Fig. 2.** Anodic and cathodic polarization curves of MS in solutions of 1 M HCl without and with different concentrations of DE

## 3.2. PDP investigation

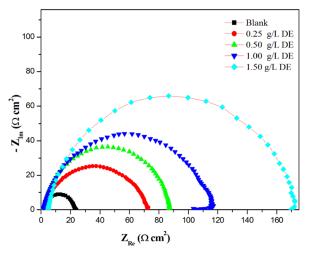
PDP of MS in 1 M HCl were obtained without and with different concentrations of each inhibitor (Fig. 1 and 2). Then, the outputted parameters of PDP analysis are given in Table 2. From Figs. 1 and 2, we noticed that both cathodic and anodic curves were translated to lower current densities with an increaser concentration of each inhibitor. Furthermore, the maximum displacement in  $E_{corr}$  value was 44.2 mV for OE and 12.7 mV for DE, which indicates that the aqueous extracts acts as mixed-type [22]. Additionally, we remarked that the inhibition efficiency EIPDP(%) increases with increasing of inhibitors concentration to reach its maximum values 91.02 and 85.05 % at 1.5 g/L for the OE and DE respectively. This result suggests that OE is considered a good inhibitor to limit the deterioration of MS in the acidic medium of HCl.

#### 3.3. EIS investigation

The EIS curves of the MS in 1M HCl were obtained in both the absence and presence of various concentrations of each inhibitor after an exposure time of 30 min at 308 K (Figs .3 and 4).



**Fig. 3.** Nyquist plots of MS in 1 M HCl without and with different concentrations of OE at 308 k



**Fig. 4.** Nyquist plots of MS in 1 M HCl without and with different concentrations of DE at 308 K

As exposed in Figs 3 and 4, we perceived that increase of different concentrations of inhibitors lead to the increasing of the semi-circle diameters of MS; this suggesting the prominent role of each inhibitor to block corrosion over MS metal. Moreover, the presence of a unique capacitive loop indicated that the studied adsorption is governed through a charge transfer process [23]. Moreover, the double-layer capacity values C<sub>dl</sub> calculated in with each inhibitor are lower concerning those in the blank solution. This suggests the formation of the protective film during adsorption by OE and DE molecules onto MS [24]. The EIS parameters are listed in Table 3. From this Table, we noticed that the charge transfer resistance value Rct

increases with the increase of different concentrations of inhibitors to reach its maximum values 286 and 170  $\Omega$  cm<sup>2</sup> at 1.5 g/L for the OE and DE respectively. Again, OE shows itself to be the best aqueous extract of *Aaronsohnia pubescens* similar to that obtained for I<sub>corr</sub> in this current research.

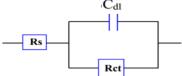


Fig. 5. The equivalent circuit used to fit the EIS experiment data

	C (g/L)	-E <sub>corr</sub> (mV/SCE)	$R_t$ ( $\Omega \ cm^2$ )	f <sub>max</sub> (Hz)	Cdl (µF cm <sup>-2</sup> )	E <sub>EIS</sub> %
1M HCl		490.1	22.00	100	73.37	
	0.25	462.9	79.00	44.64	45.25	72.15
	0.50	460.1	130.00	28.09	43.58	83.07
OE	1.00	445.8	171.72	22.32	41.52	87.18
	1.50	470.0	286.0	28.00	19.87	92.30
	0.25	497.8	74.00	31.64	67.97	70.27
	0.50	479.8	87.38	35.71	51.00	74.82
DE	1.00	477.3	115.67	35.71	38.53	80.98
	1.50	480.0	170.00	30.00	31.20	87.05

**Table 3.** EIS parameters for MS in 1M HCl without and with various concentrations of aqueous extracts OE and DE at 308 K

## 3.4. Kinetic/Activation parameters

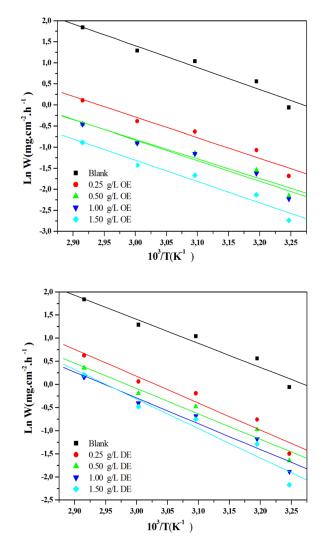
According to the Arrhenius plot Ln (W/T) versus  $10^3$ /T, we calculated the enthalpy  $\Delta H^{\circ}_a$ and entropy  $\Delta S^{\circ}_a$  energies; then the standard activation  $E^{\circ}_a$  was calculated from the plot of ln(W) against  $10^3$ /T. These parameters are associated for MS without and with inhibitors basing on the following equations: Eq.7 for  $\Delta H^{\circ}_a$  and  $\Delta S^{\circ}_a$  energies [25] and Eq. 8 for  $E^{\circ}_a$  energy [26]. The outputted parameters from the Arrhenius plot (Fig. 6 and Fig. 7) are listed in Table 4.

$$\ln\left[\frac{W}{T}\right] = \ln\left[\left(\frac{R}{N_{a}h}\right) + \frac{\Delta S_{a}^{2}}{R}\right] + \left[-\frac{\Delta H_{a}^{2}}{R T}\right] \quad (Eq.7)$$
$$\ln W = \ln A' + \left[-\frac{E_{a}^{2}}{R T}\right] \quad (Eq.8)$$

Where W, R, T, h and N<sub>a</sub> signify the corrosion rate, the universal constant of the ideal gas, the absolute temperature, the Plank constant and universal Avogadro number, respectively.

		E°a	ΔH°a	$E_{a-}\Delta H^{o}{}_{a}$	$\Delta S^{\circ}{}_{a}$
	C (g/L)	(KJ. mol <sup>-1</sup> )	(KJ. mol <sup>-1</sup> )	(KJ. mol <sup>-1</sup> )	(J. mol <sup>-1</sup> . K <sup>-1</sup> )
	1MHCl	43.35	40.65	2.70	-112.32
	0.25	40.74	38.04	2.70	-134.24
OE	0.50	38.62	35.92	2.70	-145.00
	1.00	40.69	38.00	2.70	-138.91
	1.50	42.11	39.42	2.70	-138.61
	0.25	48.29	45.59	2.71	-122.13
DE	0.50	45.66	42.96	2.70	-117.86
DE	1.00	46.35	43.65	2.70	-117.45
	1.50	52.96	50.26	2.71	-97.82

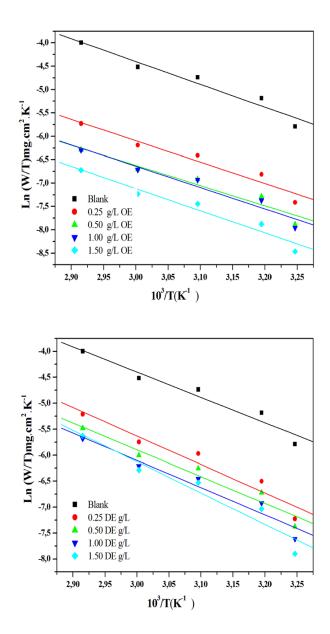
**Table 4.** Enthalpy  $\Delta H^{\circ}_{a}$  and entropy  $\Delta S^{\circ}_{a}$  energies for MS without and with various concentrations of OE and DE



**Fig. 6.** Arrhenius plots for MS corrosion rates (W) in 1M HCl in the absence and presence of different concentrations of each inhibitor

It is crystal clear from the data in Table 4 that the increase of  $E_w(\%)$  with temperature and the lower value of  $E^{\circ}_{a}$  are in favor of chemisorption for OE. Generally, the increase of  $E_w$  and the lower value of  $E^{\circ}_{a}$  of the corrosion process in an inhibitor's presence when compared to that in its absence is attributed to its chemisorption on the steel surface, while the opposite is the case with physical adsorption [27], which indicates the formation of an adsorption film of physical electrostatic nature for DE.

Additionally, the positive value of standard activation enthalpy reveals the endothermic process of corrosion phenomenon of MS [28]. However, In other words, the positive value of the standard activation entropy showed a high disorder of the inhibitors molecules during adsorption [29].



**Fig. 7.** Transition-state plots for MS corrosion rates (W) in 1 M HCl in the absence and presence of various concentrations of each inhibitor

## 3.5. Adsorption isotherm and thermodynamic parameters

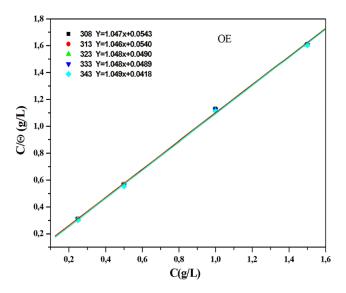
To get information about the adsorption and the surface behavior of inhibitor molecules, different models of adsorption isotherms considered. In this current study, the corresponding adsorption with different concentrations of OE and DE at different temperatures (308 to 348 K) was tested according to the following isotherms: Langmuir, Temkinand Frumkin. The findings found show that the studied adsorption obeys a Langmuir adsorption isotherm (Fig. 8 and 9) assumed by Eq. 9 [30]. As mentioned in Table 5, the linear regression factor of this isotherm model appears close to 1.

$$\frac{C}{\theta} = \frac{1}{K_{ads}} + C$$
 (Eq. 9)

where  $K_{ads}$  is adsorption coefficient. The  $K_{ads}$  values can be calculated from the intercept lines on the  $C_{inh}/\theta$  axis. The  $K_{ads}$  is linked to the standard free energy of adsorption  $\Delta G^{\circ}$  as follows (Eq. 10):

$$K_{ads} = \frac{1}{55.5} \exp\left(-\frac{\Delta G^{\circ} ads}{RT}\right)$$
(10)

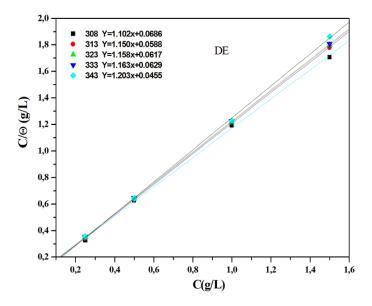
where R, T, 55.5 are gas constant, absolute temperature of experiment the molar concentration of water in solution in respectively.



**Fig. 8.** The Langmuir adsorption isotherm of OE on the MS surface in 1 M HCl at different temperatures

We can conclude from the Table 5 OE has a greater value indicating its ability to be absorbed easily and strongly [31]. Also, it is clear that, the negative values of  $\Delta G^{0}_{ads}$  suggest that the adsorption of inhibitor molecules onto MS surface is a spontaneous phenomenon. Here, the calculated  $\Delta G^{0}_{ads}$  values are ranging from -24.58 to -28.82 kJ.mol-1, This result suggests

that the studied adsorption is may be a combination of both electrostatic-adsorption and chemisorption [30].



**Fig. 9.** The Langmuir adsorption isotherm of DE on the MS surface in 1 M HCl at different temperatures

**Table 5.** Langmuir isotherm adsorption parameters for 1 M HCl/OE and DE/MS interface at various temperatures

	T(K)	R <sup>2</sup>	Kads (L. g <sup>-1</sup> )	$\Delta G^{\circ}ads$ (KJ. mol <sup>-1</sup> )
	308	0.999	18.41	-25.66
	313	0.999	18.51	-25.58
OE	323	0.999	20.40	-26.66
	333	0.999	20.49	-27.50
	343	0.999	24.39	-28.82
	308	0.999	14.70	-24.58
	313	0.999	17.24	-25.40
DE	323	0.998	16.39	-26.07
	333	0.999	16.12	-26.84
	343	0.999	22.02	-28.56

Molecule adsorption of the OE and DE at the MS surface can be attributed to the presence of electronegative elements such as oxygen and nitrogen atoms and also to the presence of  $\pi$ -electrons. The high inhibitive performance of OE is due probably to the synergic effect of their volatile especially, E-anethole (15.4%), (E)-heptadeca-10,16-dien-7-one (11.1 %), and Z-β-

Ocimene (8.6%) were identified as major constituents of the essential oil from *Aaronsohnia pubescens* (Figure 10) and non-volatile compounds. These results are coherent those of the studies previously reported in our group [10,13].

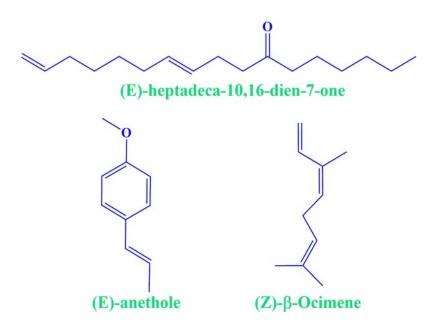


Fig. 10. The main compound structures of Aaronsohnia pubescens essential oil

## 4. CONCLUSION

In this current work, we conclude that the aqueous extracts of *Aaronsohnia pubescens* proved significant corrosion inhibition activity. PDP curves demonstrate that inhibitors acts as a mixed type inhibitor in 1 M HCl. The inhibition is accomplished by a mixture physical and chemical adsorption of the extract components on the MS surface. The aqueous extracts obeys a Lagmuir adsorption isotherm. The Nyquist diagram showed that adsorption mechanism of OE and DE are governed by a charge transfer process. Furthermore, the PDP measurements revealed that inhibitors acts as mixed type inhibitor. The high protective efficiency of OE is due to the synergic effect of their volatile and non-volatile molecules.

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