Adsorption and Inhibitory Effect of Artemisia Herba Alba Essential Oil on Corrosion of 304 Stainless Steel in Acid Medium

Brick Chaouche Souad and Haddad Ahmed

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Adsorption and Inhibitory Effect of Artemisia Herba Alba Essential Oil On corrosion of 304 stainless steel in 1 M HCl

Souad Brick chaouche[1,*], Ahmed Haddad[2]

[1.] *University of Science and Technology Houari Boumediene, BP 32 El Alia 16111 Bab Ezzouar, Algiers, Algeria*

[2.] *Research Center in Industrial Technologies CRTI, BP 64, Cheraga 16014, Algiers, Algeria*

Email: brick_chaouche@hotmail.com

Abstract

follow Langmuir adsorption In the present study, Artemisia herba-alba essential oil was investigated as green inhibitor for stainless steel corrosion in 1 M HCl solution. Aerial parts of Artemisia herba-alba was hydrodistilled, and the chemical analysis of essential oil extract has been studied using gas chromatography and GC-mass spectrometry. The effects of natural Artemisia oil on the corrosion of stainless steel were studied by the measurements of weight loss, electrochemical, and EIS polarisation. The results revealed an increase in inhibition efficiency through increasing the oil concentration. The oil compounds act as mixed-type inhibitors and adsorb by physisorption, isotherm.

Keywords: *corrosion, green inhibitor, stainless steel, acid solution, natural plant, Artemisia herba alba.*

1. Introduction

The research in the field of corrosion protection of materials is presently focused on biodegradable products known as green inhibitors, to take into account the new environmental protection guidelines [1]. For this purpose, natural products can be considered as the inexhaustible source. Recently, many plant extracts have been reported in the literature as effective corrosion inhibitors for materials in aggressive environments [2-10]. Essential oils and plant extracts have become important as an environmentally friendly, and renewable source for a wide range of inhibitors in addition, to being cheap and eco-friend.

In this work, Artemisia herba-alba essential oil was tested as a corrosion inhibitor of stainless steel in acidic environment. This plant is a greenish-silver perennial herb, belongs to the daisy family Asteraceae. It grows wild in arid areas of the Mediterranean basin. The vegetative growth of this plant takes place in the autumn; the flowering starts from September to December and basically develops at the end of the summer with many basal, erect and leafy stems covered by woolly hairs [11]. This plant is known as a medicinal and an aromatic plant. In addition, it is an effective inhibitor of foodborne pathogens, as a natural antioxidant, and is used in potential pharmaceutical applications [12-13].

In Algeria, this plant generally known as the white wormwood in Arabic as “Chih” and in France as “Armoise blanche”. In folk medicine, was used for treatment of colds, coughing, intestinal

disturbances, as antidiabetic agent for bronchitis, diarrhea, neuralgias and hypertension. Many researchers have reported various biological and/or pharmacological activities of Artimisia herba-alba essential oil as an antibacterial (bacteria and fungi), antileshmanial, anthelmintic and antispasmodic agent [12-15]. Considerable work on the composition of the essential oils of Artemisia herba-alba is reported in literature. these studies have shown that this oil is rich in β-thujone (37 et 43 %), trans-sabinyl acetate (17- 46 %), and α-thujone (10 %), it also contains: 1,8-cineole (3%) and chrysanthenone (2.5%) in small quantities [16-19].
In this article, the Artemisia herba-alba oil from Algeria was obtained by hydrodistillation and its chemical composition was investigated by capillary GC and GC/MS. Gravimetric techniques, electrochemical polarisation, and EIS measurements are applied to study the ability of Artemisia herba-alba essential oil to inhibit the corrosion of stainless steel in 1M HCl.

2. Experimental Work

2.1. Inhibitors

2.1.1. Plant material and Essential oil isolation

The aerial parts of *Artemisia herba-alba* were collected in march 2019 (stems, leaves). The entire plants were dried in the shade and stored in the laboratory at room temperature. The essential oil was extracted by hydro-distillation (2 h) from the plant using a Clevenger type distillation apparatus (1000 mL of water for 150 g plant material). A yellow oil was obtained in a yield of 2% (w/w) according to the dry material, and sodium sulfate (Na_2SO_4) was added as a drying agent to the decanted essential oil. After extraction, the essential oil was stored in a brown glass bottle tightly closed and maintained at a temperature of 4 ◦C until used.

2.1.2. Components identification.

The analysis of essential oil from *Artemisia herba-alba* was carried out by Gas chromatography analysis (GC-FID) and Gas chromatography mass spectrometry (CG/SM). The identification of the essential oil constituents was based on the comparison of their retention index (RI), calculated relative to the retention times of a series of C-5 to C-30 n-alkanes, with linear interpolation, with those of our own library of authentic compounds or literature data [15-17].

2.2. Preparation of solutions

The corrosive solution used in this study was 1M HCl. The test solutions were freshly prepared by the dilution of analytical grade 37% HCl with distilled water up to the optimum inhibitor concentration. and the solution in the absence of inhibitor was taken as blank for comparison purposes. Concentrations of essential oils were 0.05, 0.1, 0.2, 0.3 and 0.4 g/L. Experiments were conducted on several occasions to ensure reproducibility.

2.3. Materials

The chemical composition of the stainless steel 304 used in this study is given in Table 1

Table 1 – Chemical composition of the 304 stainless steel (wt %)

C	Si	Mn	P	S	Cr	Ni
<0.7	<1.00	<2.00	<0.045	<0.015	17.0-19.5	8.0-10.5

2.4. Weight loss measurements

The gravimetric test is based on the immersion of the stainless steel samples, in 200 ml of a 1M HCl solution containing the inhibitor (Artemisia herba-alba oil) at different concentrations. The stainless steel specimens used have a rectangular form (length = 2 cm, width = 1 cm, thickness = 0.05 cm). After 24 hours, the specimen was taken out, washed, dried. Then we carried out weight loss measurements of samples over time. Use of weight loss as a measure of corrosion requires making the assumption that all weight loss has been due to generalized corrosion and not localized pitting.

2.4. Electrochemical Measurements.

The electrochemical tests were carried out in a conventional three-electrode cell with a platinum counter electrode, a saturated calomel reference electrode and 304 stainless steel sample as a working electrode. In all measurements, the sample was covered by an inert resin in order to get an exposed area of 1 cm². Polishing was carried out using successively finer grade of emery papers (600–1200 grade).

The experimental apparatus used for electrochemical studies was the autolab PGSTAT302N potentiostat, monitored by a PC computer and NOVA 2.0 software. The potential scan was carried out at a rate of 10 mV/min over the potential range from -800 to -200 mV/SCE. Electrochemical impedance spectroscopy measurements (EIS) were conducted in the frequency range of 100 kHz-100 mHz, with an amplitude signal of 10 mV peak to peak. The impedance diagrams are given in the Nyquist representation. In this study, all electrochemical measurements are repeated three times to ensure the reproducibility of the tests.

3. Results and Discussion

3.1. Essential oil composition

The identified compounds are listed in Table 2, the chemical composition of essential oil was characterized by 27 compounds, which accounted for 95,2% of the total oil. The essential oil was characterized by high amounts of β-thujone 12 (38.9%). The other major components were camphor 14 (5.6%), 1,8-cineole 8 (7.1%) and α-thujone 11 (5.4%). The 22 other compounds are reported in low amounts in this essential oil.

Table 2. Chemical composition of Artemisia essential oil.

N°	Components	%
1	α-Thujene	0.4
2	α-Pinene	10.9
3	Camphene	0.7
4	Sabinene	6.8
5	α-Terpinene	0.8
6	p-Cymene	1.8
7	1,8-Cineole	7.1
8	γ-terpinene	0.5
9	(E)-Sabinene hydrate	0.7
10	α-Thujone	5.4
11	β-Thujone	38.9
12	transp-Menth-2-en-1-ol	0.6
13	Camphor	5.6
14	trans-Sabinol	1.0
15	Borneol	1.5
16	3-Thujen-10-al	0.3
17	Terpinen-4-ol	3.5
18	Myrtenal	0.2
19	α-Terpineol	0.7
20	Myrtenol	0.3
21	Piperitone	0.2
22	(E)-β-Caryophyllene	0.6
23	Germacrene D	1.4
24	Bicyclogermacrene	0.8
25	δ-Cadinene	0.3
26	Ledol	0.5
27	τ-Muurolol	2.9

The oil was dominated by oxygenated monoterpenes (64.0%) followed by monoterpene hydrocarbons (10.0%). The sesquiterpene hydrocarbons and oxygenated sesquiterpenes accounted only for 3.5% and 6.5% of the total essential oil, respectively.

It should be noted that several studies have been published on the chemical composition of *Artemisia herba-alba*. The main compounds in the essential oil of *Artemisia herba-alba* from Algeria were α -thujone (31.50 – 41.23%) and camphor (16.20 – 24.58%) [18]. Dob et al. are found that camphor (19.4%) and trans-pinocarveol (16.9%) are the major component, in the same country [19]. Dahmani-Hamzaoui and Baaliouamer observed that the main compound found were camphor (49.3%) and 1,8-cineole (13.4%), which are the same main compounds found in this study [20]. in essential oil of Artemisia herba-alba from Tunisia α-thujone was observed as the dominant constituent [17], followed by α-thujone, camphor, chrysanthenone or trans-sabinyl acetate. several studies have been published on the chemical composition of Artemisia herba-alba from Maroc [21-23], and showed that the composition of the essential oil was rich in α-thujone (60%).
In general, this variation in the chemical composition can be understandable according to exogenous factors: the period of sunshine, the nature and the composition of the ground [24]

3.2. Effect of inhibitor

3.2.1. weight loss measurements

The corrosion rate of stainless steel coupons and the inhibition efficiency in the absence and presence of different concentrations of Artemisia oil are given in table 3. The corrosion rate and inhibition efficiency (IE%) was determined by using the following Equation (1) and (2):

$$Vcorr = \frac{\Delta m}{S.t} \qquad eq.1$$

Where Δm is the weight loss, s is the specimen surface and t the immersion time.

$$IE\% = \frac{\text{Vcorr}-\text{Vinh}}{\text{Vcorr}} \qquad eq.2$$

Where V_{corr} and V_{inh} are the values of the corrosion rate in the absence and presence of inhibitor respectively.

As can be seen, the corrosion rate of 304 stainless steel in 1M HCl solution decreased with the increase of inhibitor concentration. The inhibitory efficiency increases with the concentration of the inhibitor to attain maximum values of 82 %. this increase in efficiency is due to the formation of a protective film originated by the strong adsorption of the inhibitor molecules at the metal/solution interface [25].

Table 3. Corrosion rate and inhibitor efficiency in the absence and presence of Artemisia oil after 24 h of immersion.

C (g/l)	**IE (%)**	**V_{corr} ($mg.cm^{-2}. h^{-1}$)**
blank	-	0.6539
0.05	27.47	0.4742
0.1	41.89	0.3799
0.2	57.7	0.2770
0.3	62.36	0.2477
0.4	82.166	0.1297

3.2.2. Potentiodynamic Polarization Studies.

Figure 1 shows the potentiodynamic polarization curves recorded with a 304 stainless steel electrode immersed in 1M HCl solution, in the absence and presence of different concentrations of Artemisia herba-alba oil.

It is apparent from Figure 1, that the polarization curves were shifted into more negative values of potential. And the cathodic and anodic current densities toward lower values with the increase of inhibitor concentration. However, it suggests that this decrease of current densities is due to the formation of a layer on the stainless steel surface by adsorption of the inhibitor molecules. This adsorption phenomenon is due to an interaction between the components of the chemical composition of the inhibitor with material surface [26]

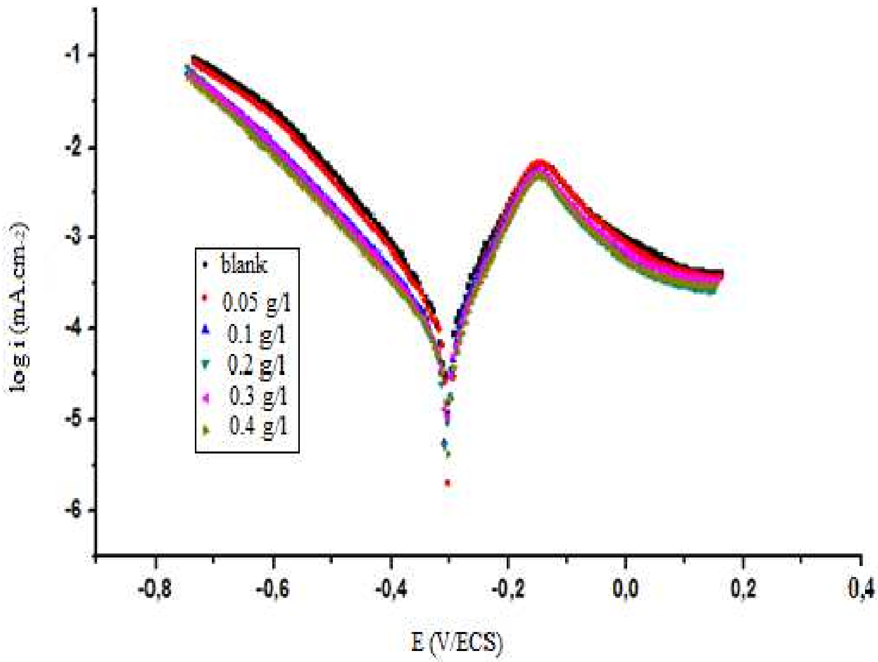

Fig. 1. Potentiodynamic polarization curves for stainless steel after 24 h of immersion in 1 M HCl with and without Artemisia oil

The polarization parameters, the corrosion potential (Ecorr), the cathodic and anodic Tafel slopes (bc and ba), and the corrosion current densities (icorr), obtained by extrapolating the Tafel lines to Ecorr, are indicated in Table 4. From the table, it is seen that both the anodic and cathodic Tafel constants are varied with increasing Artemisia oil concentration, which indicates the influence of the Artemisia herba-alba oil on the cathodic and anodic reactions, but the cathodic curves are more affected. On the other hand, we note that the addition of the inhibitor did not modify the corrosion potential values (Ecorr). This behavior indicates that Artemisia oil acts as a mixed-type inhibitor [27-29]. Further, these results show that increasing the essential oil concentration decreased the corrosion current density (icorr), therefore decreasing the rate of the electrochemical reaction. These results were used to calculate the inhibitory efficiency (%)EI by using corrosion current density as follows [27]:

$$IE\% = \frac{i0\ corr - icorr}{i0corr} * 100 \qquad \text{eq. 3}$$

Where i_{corr} and i_{0corr} are the corrosion current density value with and without inhibitor, respectively. The inhibitory efficiency increased with the concentration of the inhibitor until reaching the maximum value of 83% at 0.4 g. L^{-1} of Artemisia herba-alba oil. The results are in good agreement with the weight loss measurements.

Table 4. Electrochemical parameters of stainless steel in 1M HCl in the presence of different concentrations of inhibitor.

C (g/l)	E_{corr} (mV/ECS)	ba (mV/dec)	-bc (mV/dec)	I_{cor} (µA.cm-2)	IE (%)
Blanc	-321	295.4	351.8	56 .27	-
0.05	-328	430.3	941.6	40.81	27.47
0.1	-332	271.2	398.3	32.69	41.89
0.2	-327	353	325.7	23.89	57.71
0.3	-333	346.2	254.8	20.61	62.36
0.4	-325	332.4	241.3	10.16	83.17

3.2.3. EIS Studies

A better understanding of the characteristic features and kinetics of the electrochemical reactions that occur at the electrode surface was attained through EIS measurement. The impedance responses without and with different concentrations of Artemisia oil in 1M HCl solution were studied by EIS and are expressed in terms of Nyquist plot as shown in Figure 2. This indicates that the addition of Artemisia oil increases the corrosion resistance of the metal due to the increased amount adsorbed on the metallic substrate. and that, the corrosion of stainless steel in HCl solution is mainly controlled by a charge-transfer process.

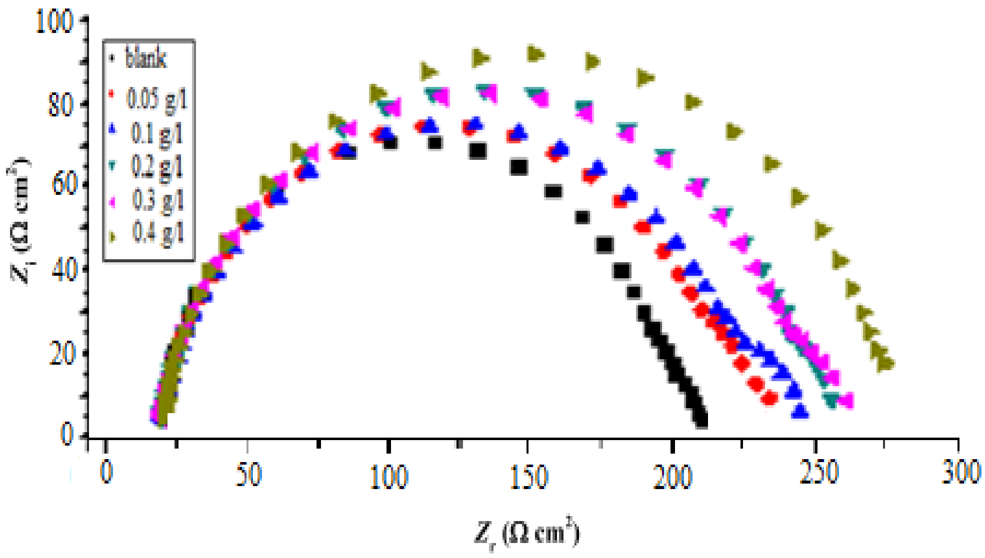

Figure 2. Nyquist plots of stainless steel in absence and presence of different concentrations of Artemisia oil in 1M HCL.

The EIS parameters derived from Figure 2 have been simulated by the proposed equivalent electrical circuit presented in Figure 3. The equivalent circuit was established on the basis of a general knowledge of the physical events which can occur at the interface [30]. The different parameters from electrochemical EIS measurements, such as The charge-transfer resistance (R_{ct}), double-layer capacitance (C_{dl}), and inhibition efficiency (EI%) are summarized in Table 5. The inhibition efficiency (EI%) was calculated according to the following equation:

$$IE\% = \frac{Rct - Rct0}{Rct} * 100 \qquad \text{eq. 4}$$

Where R_{ct} and R_{ct0} are the charge-transfer resistance values with and without inhibitor, respectively.

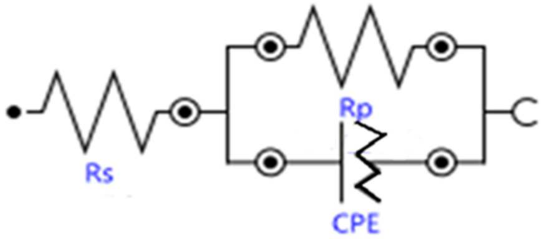

Figure 3 . Equivalent circuit for the metal–solution interface.

Table 5. Corrosion parameters obtained by EIS measurements in 1M HCL at various concentrations of inhibitor.

Co (g/L)	**R_{ct} (Ω.cm^{-2})**	**C_{dl} (μf/cm^{-2})**	**E%**
Blank	178.26	89.326	-
0.05	194.80	51.576	27.50
0.1	200.10	50.209	42.20
0.2	218.15	36.583	57.69
0.3	220.69	36.162	63.60
0.4	249.04	32.045	81.26

The data in Table 5 shows that R_{ct} values increase significantly with the increase of Artemisia oil concentration. This increase is due to the gradual increase of inhibitor molecules on the metal surface and causes a decrease in the number of active sites necessary for the corrosion process [31-32]. As the IE% is directly proportional to R_{ct}, it also increases as the Artemisia oil concentration increases, and the maximum value: 81.26 % was obtained in the case of 4 g L^{-1}. The double-layer capacitance (C_{dl}) decreases with an increase in the concentration of the Artemisia oil, signifying that a higher presence of inhibitor blocks the charge transfer process and prevents the destruction of the metal [33].

The values of the inhibition efficiencies obtained by EIS measurements are in good agreement with weight loss measurements and potentiodynamic polarization, and suggest once again, that this Artemisia herba-alba plant extract could serve as an effective corrosion inhibitor.

3.2. Adsorption Isotherm and Thermodynamic Calculations.

The efficiency of the Artemisia oil as a successful corrosion inhibitor principally depends on its adsorption ability on the stainless steel surface. So it's important to know the mode of adsorption that can give detailed information on the exchanges between compounds of inhibitor and metal surface [34,35]. In this work, the best correlation between the isotherm functions and experimental results was achieved using the Langmuir adsorption isotherm (fig.4), given by eq. 5. The degree of surface coverage (θ) was obtained from the weight loss data using eq.6.

$$\frac{C}{\theta} = \frac{1}{Kads} + C \qquad \text{eq. 5}$$

Where Kads (L.g-1) is the equilibrium constant for adsorption process.

$$\theta = \frac{Vcorr - Vinh}{Vcorr} \qquad \text{eq. 6}$$

Where V_{corr} and V_{inh} are the values of the corrosion rate in the absence and presence of inhibitor respectively.

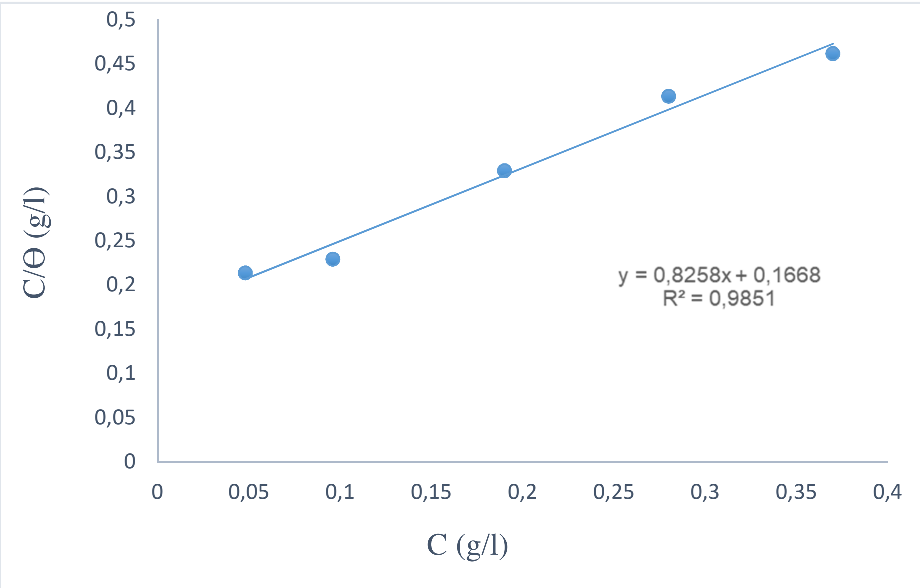

Figure 4. Langmuir adsorption isotherm plot for Artemisia oil.

The form of this curve shows that the plot of C/θ versus C is linear, both slope value and linear correlation coefficient (R) are close to 1. The model of adsorption mechanism explains the inhibitive action of Artemisia herba-alba oil on the stainless steel corrosion in an acid solution acting by the formation of a monolayer barrier of active compounds present in the oil at the surface. The value of Kads which is obtained by extrapolating the line of the Langmuir adsorption isotherm with respect to the C/θ axis is of the order of 10.92 l/g. Furthermore, the adsorption constant K is related to the standard free adsorption energy (G°ads) by the following equation 7:

$$K = \frac{1}{55,5} \exp\left(\frac{-\Delta G^\circ_{ads}}{RT}\right) \qquad \text{eq.7}$$

The values of the standard free energy of adsorption ΔG°ads calculated from equation 7 is approximately -15.98 $kJ.mol^{-1}.K^{-1}$ after the addition of the inhibitor. It is well known that The negative sign of ΔG°ads indicate a physisorption of Artemisia herba alba on the stainless steel may occur, and ensures the spontaneity of the adsorption process and stability of the adsorbed layer on metal surface [36-38]

4. Conclusion

Good results were obtained with the different methods employed in this study. The weight loss measurements, Potentiodynamic Polarization and EIS Studies showed that Artemisia herba-alba can be a good candidate for stainless steel corrosion protection in acid medium.

The results obtained showed that the essential oil of Artemisia reduces the corrosion rate of stainless steel in HCl solution. And inhibition efficiency increase with oil centration, maximum value of inhibition efficiency (83%) was reached at 0.4 $g.L^{-1}$. This indicates that the addition of Artemisia oil increases the corrosion resistance of the metal due to the increased amount adsorbed on the metallic surface. The studies of the adsorption process showed a Langmuir adsorption process with a physisorption aspect, established by the value of the adsorption energy (ΔG°ads = -15.71 $kJ.mol^{-1}$). the corrosion of stainless steel in HCl solution is mainly controlled by a charge-transfer process, and the oil of Artemisia behaves as a mixed type inhibitor.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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