Environmentally friendly inhibitor of the corrosion of mild steel: Commercial oil of *Eucalyptus*

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Abstract

A study assessing plant extracts as green corrosion inhibitors is important because of the potential cost effective and environmental benefits. This study employed a strategy to evaluate the effectiveness these molecules against corrosion of mild steel in acid media. Many plant extracts have been used as corrosion inhibitors of iron or steel in acidic media. This research aims to examine the effects of commercial oil of *Eucalyptus* on inhibition of corrosion with mild steel in hydrochloric acid by means of gravimetric, potentiodynamic polarization and electrochemical impedance spectroscopic measurements. The results of the polarization curves show that the corrosion current density decreases from 3618 $\mu A/cm^2$ to 87 $\mu A/cm^2$ with the addition of the *Eucalyptus* inhibitor. The charge transfer resistance increases from 17 $Ohm{\cdot}cm^2$ to 185 $Ohm{\cdot}cm^2$ in the electrochemical impedance spectrum after the addition of the oil *Eucalyptus* inhibitor in the more concentrated solutions It is found that the adsorption of our green inhibitor on steel surface obeys the Langmuir adsorption isotherm equation. The inhibition efficiency was found to be increased with increasing immersion time of samples in the solution. The polarization plots indicate that the studied *Eucalyptus* inhibitor affects both the anodic metal dissolution, and the catholic hydrogen evolution acting as a mixed type inhibitor.

Keywords: *Eucalyptus, commercial oil, inhibition, corrosion, mild steel, green inhibitor*.

Received: June 30, 2019. Published: July 31, 2019 doi: 10.17675/2305-6894-2019-8-3-8

I. Introduction

Corrosion is a natural process, which converts a refined metal to a more chemically-stable form, such as its oxide, hydroxide, or sulfide. It is the gradual destruction of materials (usually metals) by chemical and/or electrochemical reaction with their environment. Corrosion engineering is the field dedicated to controlling and stopping corrosion [1–5].

Even though most of the materials such as ceramics and glass also decay, usually, the term corrosion is concerned with metals. Corrosion is a natural process, spontaneous and thermodynamically stable process favored by nature [6–10].

So no one can prevent corrosion. However the rate of corrosion can be controlled, just like postponing death.

There are many methods to control corrosion such the use of inhibitors in small quantities is a common process. Once organic compounds were used as corrosion inhibitors effectively. Yet, environmental scientists point out the health hazards caused by organic compounds. Hence corrosion scientists go for environmentally friendly non-toxic natural products extracts as corrosion inhibitors [11–13].

Many parts of plant materials such as bark, seeds, flowers and leaves can be used as corrosion inhibitors. Several research studies have been published on this subject [14–16].

In the meaning of expanding green and environmentally friendly inhibitors and further research into the activity of plants oils in different fields, we report the applicability of oil *Eucalyptus* as an efficient and green inhibitor for mild steel in HCl solution by using electrochemical impedance spectroscopy (EIS), gravimetric and polarization resistance analyses.

In the present study, the inhibitive effect of oil *Eucalyptus* on corrosion of mild steel in hydrochloric acid solution (1 M HCl) was investigated using weight loss, potentiodynamic polarization and electrochemical impedance spectroscopy (EIS) methods.

II. Experimental

II.1. Plant material

Kingdom: Plantae

There are more than 800 species of eucalyptus and most are native to Australia; a very small number are found in adjacent areas of New Guinea and Indonesia. One species, *Eucalyptus deglupta*, ranges as far north as the Philippines. Of the 15 species found outside Australia, just nine are exclusively non-Australian. Species of eucalyptus are cultivated widely in the tropical and temperate world, including the Americas, Europe, Africa, the Mediterranean Basin, the Middle East, China and India. However, the range over which many eucalypts can be planted in the temperate zone is constrained by their limited cold tolerance.

II.2. Anticorrosion effects

II.2.1. Materials

Tests were performed on cold rolled steel (CRS) with a composition of 0.09% P; 0.38% Si; 0.01% Al; 0.05% Mn; 0.21% C; 0.05% S and the remainder iron. The materials were polished with emery paper up to grade 1200, washed thoroughly with doubly-distilled water, degreased with AR grade ethanol and acetone, and dried at room temperature.

MS samples of 1.0×1.0×0.5 cm and MS powder were used for weight loss studies. For electrochemical studies, specimens with an exposed area of 1 cm^2 were used. These specimens were degreased ultrasonically with 2-propanol and polished mechanically with different grades of emery paper to obtain very smooth surfaces.

II.2.2. Preparation of the solutions

The test solutions were prepared by dilution with analytical grade 37% HCl with distilled water.

The inhibitor was dissolved in the acid solution at the required concentrations (mol/L) and the solution in the absence of the inhibitor was taken as blank for comparison purposes.

Experiments were conducted on several occasions to ensure reproducibility. Concentrations of inhibitor were 0.25, 0.5, and 1 g/L.

II.2.3. Electrochemical measurements

Testing of potential free and spectroscopy electrochemical impedance (EIS) was performed using a potentiostat PGZ 301 (Radiometer Analytical) controlled by the VoltaMaster 4 software. This assembly comprises three electrodes: steel as a working electrode (ET), platinum as a counter electrode (CE), and Ag/AgCl as a reference electrode.

III. Results and discussion

III.1. Oil composition of Eucalyptus

The GC-MS result of *Eucalyptus* oil showed that the sample consist of 22 components, representing 98.80% of the total components identified (Table 1). The major phytochemicalin the oil were: palmitic acid (29.00%), oleic acid (10.00%), *E*,*E*,*E*-*α*-springene (9.00%), 2-ethenyl-2,5-dimethyl-4-hexen-1-ol (8.00%), 2,4-dimethylheptane (6.00%), hexahydrofarnesyl acetone (5.0%), geranylbutanoate (4.00%), farnesol (4.00%), geranyl-geraniol (4.00%) and *trans*-2-methyl-2-(4-methyl-3-pentenyl)-cyclopropanecarbox-aldehyde (4.00%).

From the GC-MS analysis, it appears that the composition of commercial oil of *Eucalyptus*.

Table 1: Chemical composition of *Eucalyptus* oil.

Compound	Percentage Composition
2,4-Dimethylheptane	6.0
α-Pinene	4.0
p-Cymene	0.4
1,8-Cineole	0.5
Phthalic acid, di(1-hexen-5-yl) ester	1.0
Palmitic acid	29.0
Oleic acid	10.0
Geranyl butanoate	4.0
Hexahydrofarnesyl acetone	5.0
2,4-Dibromopentane	0.4
2,4-Dimethyl-2,4-heptadiene	0.05
Levomenol	1.0
E,*E*,*E*-α-Springene	9.0
2-Ethenyl-2,5-dimethyl-4-hexen-1-ol	8.0
5-Bromo-*n*-pentanol-cyclohexyl ether	2.0
threo-2,3-Dibromopentane	2.0
Farnesol	4.0
Lavandulol	1.0
1,1'-Bicyclooctyl	1.0
Geranylgeraniol	4.0
(*E*,*E*)-Geranyl linalool	2.0
trans-2-Methyl-2-(4-methyl-3-pentenyl)-cyclopropanecarboxaldehyde	4.0
Percentage Total	98.8

III.2. Weight loss study

III.2.1. Effect of inhibitor concentration

Table 2 shows the results obtained from weight loss measurements for mild steel in 1.0 M HCl solutions in the absence and presence of different concentrations of inhibitor (Oil Eucalyptus). It has been observed from the results that the ηWL% of Oil *Eucalyptus* increases from 86% to 93% with the increase in inhibitor concentration from 0.25 to 1 g/L. Indeed, corrosion rate values of MS decreases from 0.112 to 0.061 $mg/cm^2 \cdot h$ on the addition of 0.25 to 1 g/L of Oil *Eucalyptus*. The increase in efficiency from 86% to 93% may be due to the blocking effect of the surface by both adsorption and film formation mechanisms, which decreases the effective area of corrosion attack.

The results confirm that Oil *Eucalyptus* is an efficient corrosion inhibitor eco-friendly, which gives efficiency values as high as 93% in temperature 308 K. The inhibiting performance exhibited by the Commercial oil of *Eucalyptus* may be due to the presence of the Compounds organic (see Table 1) which makes it adsorb quickly on the mild steel surface, thus forming an insoluble stable film on the surface of the mild steel.

Table 2: Weight loss values of various concentrations of Oil Eucalyptus in 1.0 M HCl solution.

Concentrations (g/L)	C_R ($mg/cm^2 \cdot h$)	η_{WL} (%)	θ
HCl (1 M)	0.82	–	–
0.25 g/L	0.112	86	0.86
0.50 g/L	0.097	88	0.88
1.00 g/L	0.061	93	0.93

III.3. Adsorption isotherm

Metal surface providing us several information about the adsorption mechanism of the inhibitors on the surface by studying the relationship between the concentration and the surface coverage. Many isotherms are employed to fit the experimental data such as the Langmuir, Freundlich, Temkin, Dubinin–Radushkevich, Frumkin, and BET (Brunauer, Emmett and Teller) isotherms.

It is found that the adsorption of our studied green inhibitor on steel surface obeys the Langmuir adsorption isotherm equation [17]:

$$\frac{C}{\theta} = \frac{1}{K} + C \tag{1}$$

Where C is the concentration of inhibitor, K the adsorption equilibrium constant, and θ is the surface coverage.

Plots of C/θ against C yield straight lines as shown in Figure 1, and the corresponding linear regression parameters are listed in Table 3. Both linear correlation coefficient (r) and slope are close to 1, indicating the adsorption of commercial oil of *Eucalyptus* green inhibitor on mild steel surface obeys Langmuir adsorption isotherm.

Furthermore, the adsorption equilibrium constant (K) is related to the standard free energy ΔG^0 by the following equation:

$$\Delta G^0_{\text{ads}} = -RT\ln(55.5K) \quad (2)$$

Where R is the gas constant ($8.314\ \text{J}\cdot\text{K}^{-1}\text{mol}^{-1}$), T the absolute temperature (K), and the value 55.5 is the concentration of water in the solution.

Generally, at values of ΔG^0_{ads} up to -20 kJ/mol, the types of adsorption were regarded as physisorption, the inhibition acts due to the electrostatic interactions between the charged molecules and the charged metal, while values around -40 kJ/mol or smaller are associated with chemisorption as a result of sharing or transfer of electrons from organic molecules to the metal surface to form a coordinate type of bond (chemisorption) [18, 19].

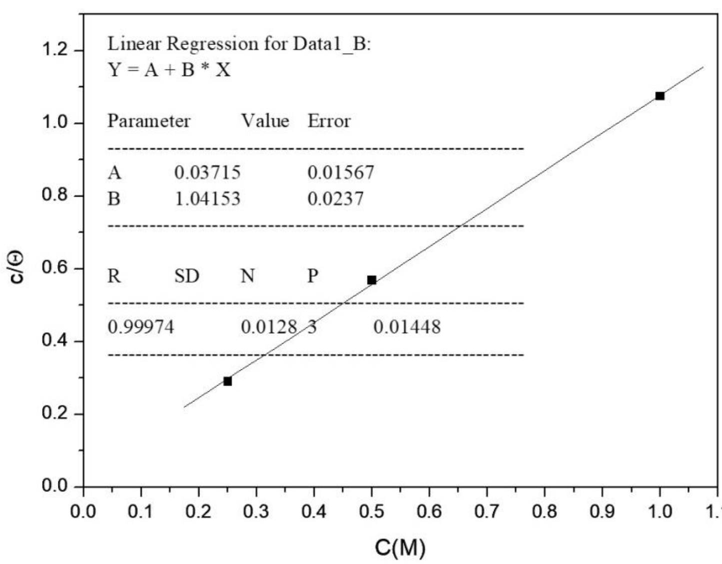

Figure 1. The Langmuir adsorption isotherm plots for the adsorption of Commercial oil of *Eucalyptus* in 1.0 M HCl on the surface of mild steel.

Then according to Equation (2), we calculated the $\Delta G^0_{\text{ads}} = -18.71$ kJ/mol. Therefore it can be concluded that the adsorption of the Commercial oil of *Eucalyptus* on the mild steel surface is mainly the physisorption.

Table 3. Thermodynamics parameters for adsorption off inhibitor Commercial oil of *Eucalyptus* on the mild steel surface in 1 M HCl.

Inhibitor	Linear correlation	Slope	*K*	ΔG^0 ($kJ \cdot mol^{-1}$)
Commercial oil of *Eucalyptus*	0.999	1.041	26.91	–18.71

III.4. Influence of temperature

To investigate the mechanism of inhibition and to determine the thermodynamic functions of the corrosion process, gravimetric test of mild steel in 1 M HCl were determined at various temperatures (318–348 K) in the absence and presence of 10^{-3} M of Commercial oil of *Eucalyptus*.

The effect of temperature on the inhibition efficiency of the Commercial oil of *Eucalyptus* is shown in Table 4. An increase in temperature from 318–348 K resulted in an increase in the corrosion rate, probably as a result of desorption of inhibitor molecules from the metal surface [20].

Table 4. Weight loss values of Commercial oil of *Eucalyptus* at various temperatures in 1.0 M HCl solution.

Temperature (K)	Inhibitor	C_R ($mg/cm^2 \cdot h$)	η_{WL} (%)
318	HCl 1 M	2.2541	–
	Commercial oil of *Eucalyptus*	0.4403	80
328	HCl 1 M	4.2581	
	Commercial oil of *Eucalyptus*	1.0054	76
338	HCl 1 M	7.189	
	Commercial oil of *Eucalyptus*	2.2723	68
348	HCl 1 M	9.9806	
	Commercial oil of *Eucalyptus*	3.5878	64

The effect of temperature on the corrosion inhibition efficiency of the Commercial oil of Eucalyptus can be best represented by Arrhenius equation which represents the natural logarithm of corrosion rate ($\ln C_R$) as a linear function of $1/T$:

$$\ln C_R = (E_a / RT) + A \quad (3)$$

Where C_R is the corrosion rate, E_a is the apparent activation energy, and A is the preexponential factor. The Arrhenius plots of $\ln C_R$ *versus* $1/T$ for the blank and optimum concentration of inhibitor Commercial oil of *Eucalyptus* give a straight line and a slope equal to $–E_a/R$ shown in Figure 2, from which the values of E_a for the inhibited corrosion reaction of mild steel have been calculated and recorded in Table 5.

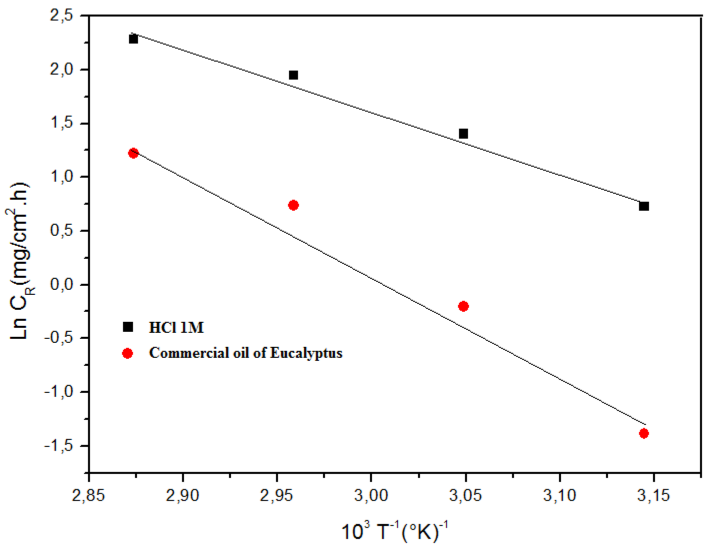

Figure 2. Arrhenius plots for mild steel corrosion rates Ln(C_R) *versus* $1/T$ in 1.0 M HCl in absence and in presence of optimum concentration of inhibitor commercial oil of *Eucalyptus*.

In 1.0 M HCl solution, the addition of inhibitor (Commercial oil of *Eucalyptus*) leads to an increase in the apparent activation energy to value greater than that of the uninhibited solution. The results show that the addition of inhibitor decreases metal dissolution in 1.0 M HCl medium. On the other hand, the increase in the apparent activation energy may be interpreted as physical adsorption that occurs in the first stage [21, 22].

Table 5. The values of activation parameters for mild steel in 1.0 M HCl in the absence and presence of 10^{-3} M of inhibitor commercial oil of *Eucalyptus*.

Inhibitor	E_a (J/mol)
HCl 1 M	48.13
Commercial oil of *Eucalyptus*	80.89

III.5. Potentiodynamic polarization curves

The polarization curves of the mild steel in 1 M HCl in the absence and presence of inhibitor commercial oil of *Eucalyptus* at different concentrations at 308 K are shown in Figure 3. Corrosion potential (E_{corr}), corrosion current (I_{corr}), Tafel cathodic slopes (β_c) and percentage inhibition efficiency (E %) are shown in Table 6.

The relationship determines Inhibition efficiency (E %):

$$E(\%) = \frac{I^0_{corr} - I_{corr}}{I^0_{corr}} \times 100 \tag{4}$$

Where I^0_{corr} and I_{corr} are uninhibited and inhibited corrosion current densities, respectively. Under the experimental conditions performed, the cathodic branch represents the hydrogen evolution reaction, while the anodic branch represents the iron dissolution reaction.

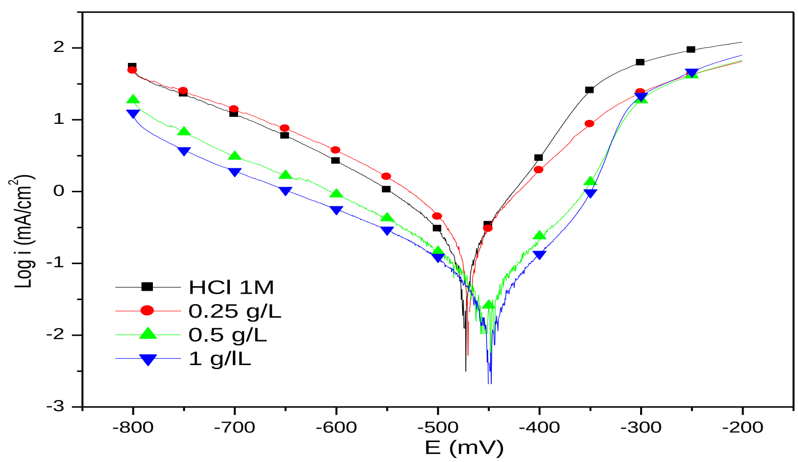

Figure 3. Tafel polarization curves in in HCl 1 M with and without inhibitor commercial oil of Eucalyptus at different concentrations.

Table 6. Corrosion parameters obtained by electrochemical measurements for mild steel in 1M HCl at various concentrations of inhibitor commercial oil of Eucalyptus.

Inhibitor concentration	$-E_{corr}$ (V)	$-\beta_c$ (mV/dec)	β_a (mV/dec)	I_{corr} (μA/cm^2)	E (%)
HCl 1 M	0.46	149	61	3618	–
0.25 g/L	0.47	165	89	649	82
0.5 g/L	0.44	170	76	394	89
1 g/L	0.43	193	60	167	95

The geometric blocking effect of the adsorbed inhibitory species, the blocking effect of active sites by the adsorbed inhibitory species and the electro-catalytic effect of the inhibitor or its reaction products, are the three classifications of inhibition modes Inhibitors [23].

The first mode of the inhibitory effect, comes from the reduction of the reaction zone on the surface of the corrosive metal, whereas the other two modes are due to changes in the mean energy barriers of activation of the reactions anodic and cathodic properties of the corrosion process.

The slight changes in the Tafel cathodic slope (β_c) with the addition of of inhibitor commercial oil of Eucalyptus, are due to the inhibitory action that occurred by simple blocking of the cathodic sites available on the metal surface [24, 25]. This leads to a reduction in the exposed surface area necessary for the evolution of hydrogen and the lowering of the dissolution rate with the increasing inhibitor concentration (Table 6).

III.6. Electrochemical impedance spectroscopy

In order to discuss the inhibition performance and mechanism of commercial oil of *Eucalyptus*, the EIS of mild steel specimens immersed in the absence and presence of inhibitor with various concentrations were displayed in Figure 4.

EIS is an important tool for determining, double-layer capacitance, C_{dl} and resistance to transfer Rt. The study of the change of these parameters as a function of time or with respect to other variables gives us important information on the kinetics of the corrosion process involved. The impedance measurements were carried out using AC signals of amplitude 10 mV peak to peak, the open circuit potential using Tacussel Radiometer PGZ 301 Frequency Response Analyzer in a frequency range of 100 kHz to 10 mHz.

Figure 4 show the electrochemical impedance graph for mild steel in the 1 M HCl solution in the absence and presence of various concentrations of commercial oil of *Eucalyptus*. The impedance data of the EIS experiments carried out in the absence and presence of different concentrations of the oil studied are summarized in Table 7.

The charge transfer resistance, R_t values are calculated from the difference in impedance at lower and higher frequencies, as suggested by several authors [26–31].

The inhibition efficiency E(%) was estimated from the charge transfer resistance measured, R_{ct}, using the following equation:

$$E\% = ((R_{ct} - R_{ct}^0) / R_{ct}) \times 100 \qquad (5)$$

Where R_{ct}^0 and R_{ct} are the charge-transfer resistance values in the absence and presence of the inhibitor, respectively. All electrochemical measurements were done in unstirred and deaerated solutions.

It is found that R_{ct} values increase with the increase of Commercial oil of *Eucalyptus* concentrations, indicating an insulated adsorption layer's formation.

R_{ct} values increased and C_{dl} values decreased with increasing inhibitor concentration. These results indicate a decrease in the active surface area caused by the adsorption of the inhibitors on the mild steel surface and this suggests that the corrosion process is hampered. The best result for the inhibition efficiency of inhibitor commercial oil of *Eucalyptus* was obtained at a concentration of 0.1 g/L, with a yield of 89%.

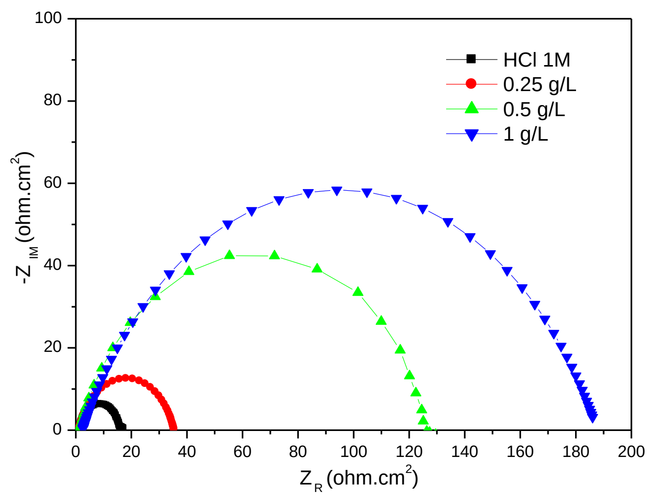

Figure 4. Nyquist plots in absence and presence of different concentrations of inhibitor commercial oil of *Eucalyptus* in HCl 1 M.

Table 7. Corrosion parameters obtained by impedance measurements for mild steel in 1 M HCl at various concentrations of commercial oil of *Eucalyptus*.

Inhibitor concentration	R_{ct} **(Ohm·cm²)**	C_{dl} **(μF/cm²)**	***E*** **(%)**
HCl 1 M	17	200	–
0.25 g/L	35	139	46
0.5 g/L	127	98	84
1 g/L	185	63	89

IV. Conclusion

Commercial oil of *Eucalyptus* exhibits a considerable inhibitive effect on mild steel corrosion in 1.0 M HCl solution. The polarization plots indicate that the studied *Eucalyptus* inhibitor affects both the anodic metal dissolution, and the cathodic hydrogen evolution acting as a mixed type inhibitor. The impedance measurements show that the charge transfer resistance (R_{ct}) and the double layer capacitance (C_{dl}) have opposite concentration relationships (R_{ct} increases, while C_{dl} decreases with the increase in the *Eucalyptus* inhibitor concentration). All the results that obtained from the electrochemical studies show an excellent agreement with the weight loss measurements.

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