

Anticorrosion Coating using *Olea* sp. Leaves Extract

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Abstract. Olive leaves extract (OLE) was evaluated as green corrosion inhibitor for stainless steel grade 316L (SS316L) in several media using scanning electron microscope (SEM), electrochemical impedance spectroscopy (EIS) and potentiodynamic polarization techniques. The Fourier Transform Infrared (FTIR) spectroscopy results reveals several active compound indicated by O-H stretch, C=O stretch, C-OH stretch and C-N stretch which can be attributed to oleuropein and hydroxytyrosol acting as the main inhibiting sources for corrosion. The results obtained also show the inhibition efficiency of OLE increase with the increase of OLE concentration. Through its inhibitive action elucidate from the electrochemical analysis, the extract was found to act as a mixed type inhibitor. Micrographs by SEM showed that the surface of steel which has been coated with 0% and 20% of OLE coating extract possess a lot of pin holes or pores while the steel with 10% of OLE coating extract shows the surface has multiple cracks. This study clearly shows the efficiency of OLE as anticorrosion coating for control of stainless steel in marine application.

1. Introduction

Corrosion is the degradation of metals resulting in an eventual failure of structures. As a ubiquitous problem face by most marine industry, complete elimination of the problem is difficult and thus a prevention methods to counter it are more practical [1]. The consequences of these inevitable threats have severely affected the operation cost causing a decrease in productivity. Reports from U.S. Department of Transportation for the period of 6 years between 1994 and 1999 suggest that approximately 25% of all reported accidents related to the pipeline system were due to corrosion [2]. Since industries depend heavily on the use of metals, practical corrosion protection of these metals has been widely studied in its usage whether in industrial application or products.

In the past, inorganic inhibitors such as phosphate and chromate are often used in marine coating due to their high efficiency [3]. However, these agents were known to cause hazard in environment due to their toxicity, non-degradable and/or bioaccumulation [4]. The first reported study which propels the beneficial property of plant extract as an inhibitor of corrosion was in 1930's when plant extract of *Chelidonium majus* (Celandine) and several others were tested for the corrosion inhibition in H₂SO₄ pickling baths [5]. Nowadays, interests in the use of an organic natural plant extract as a green inhibitor are steadily increased since these sources are much more acceptable,



economical, readily available, practical, biodegradable and convenient. Various plant extract currently studied are found containing compound mixture of oxygen, sulfur and nitrogen which has been proven to be effective against corrosion [6-8]. Several studies suggest the inhibitor action by plant extract works through the adsorption on to the steel surface. Adsorption is a process of molecules attaching directly on to the surface without penetrating into the metals itself. Without an intermediate of paint, the compound will adsorb onto the surface through the displacement of water molecules and form a thin barrier on the surface [9]. Since the use of paint is applied in the study, the adsorption process is directly responsible by the binder component of the paint.

Olea sp. or olive is a popular medicinal plant across the globe. The leaf is considered as the waste from the farming of olive grove that gathers when the olive trees prune [10]. In the region of Mediterranean or Spain where the plant are extensively grow, the olive leaf is regarded as an agricultural wastes and usually be thrown away without further beneficial use. However, recent studies show the leaf is in fact possess the potential as an excellent corrosion inhibitor due to the abundant presence of phenolic compound [11].

The fresh pulp weight of olives usually produce 1-3% of phenolic compound which are considered as highly concentrate. Phenolic acid, phenolic alcohol, flavonoid and secoiridoids are the main classes of phenolic compound found in the extract of OLE. Generally, the most abundant of phenolic alcohol are hydroxytyrosol and tyrosol. The flavonoids of OLE are flavonol glycosides which are rutin, luteolin-7-glucoside and anthocyanins. Although phenolic acids, flavonoids and phenolic alcohol occur in many plants categorized to many botanical families, secoiridoids can only be exclusively identified in plants that belong to the Oleaceae family which is focused in the research [12-15].

Nowadays, application of coating in marine industry as well as others can be virtually seen everywhere. It can be seen on the wall of machines, bridges, pipelines, and superstructures. Coatings are not useful product by themselves, but they make other products better, more durable, more attractive, safer and more saleable [16]. In general, coating are made up of four constituents i.e. binder, pigment, additive and solvent [17]. Binder exists as to hold the pigment to the surface, pigments responsible in giving the coating its color and finish, additive serve to improve or modify the property of the coating and solvents are used to control the viscosity of the coatings for acceptable application [18]. The study focus on the efficiency of OLE integrated as an additive with the use of KTH 1K Alkyd paint in inhibit the corrosion progress on SS316L. Together with the inhibitive action of the paint, the extracts are expected to significantly increase the efficiency of the coating in reducing the corrosion rate. Although this type of paint are actually standardize and suitable for mild steel, but base on the study previously show this type of paint are suitable for the integration of plant extract for corrosion analysing process.

2. Materials and Methods

2.1 Chemicals

80% ethanol, acetone, distilled water and deionized water were supplied by Universiti Malaysia Terengganu. KTH 1K Alkyd and general thinner were purchased from a local Nippon Paint Holdings Co., Ltd.

2.2 Preparation of extract

Fresh olive leaves are plucked during early morning and cleansed using distilled water to remove impurities and dirt. The leaves were dried at ambient temperature for 2 to 3 days away from the exposure of sun light. Dry leaves are later grinded to obtain the powder for further extraction. The

extraction methods to obtain OLE were done following the procedure conducted by Yateem et al. [19] with some modifications. Ten grams of olive leaves powder was weighed accurately using weighing scale and put into a conical flask. 100 ml of 80% ethanol was poured into it for maceration process. The immersed extract is left at ambient temperature for four hours. Next, the macerated solution is filtered using filter paper and evaporated using a rotary evaporator in vacuum at room temperature. The concentrated extract is stored in a refrigerator at 2-8°C before used.

2.3 FTIR Spectrum

Extract was characterized by FTIR spectroscopy for the determination of chemical functional group. Functional group of different types will absorb different characteristic of IR radiation frequencies. FTIR Spectroscopy was carried out using Thermo Nicolet Avatar 380. A small quantity of diluted extract was directly deposited onto the sample disk, creating a thin film. A spectrum was scanned in the frequency range of infrared light ranging from 4000 to 700 cm⁻¹.

2.4 Ultraviolet-Visible analysis

The absorbance of diluted extract was measured by using Lambda 25 UV-Vis spectroscopy. UV-Vis Spectroscopy is an instrument to analyze the absorption of the polymer and dyes. The 10 mm³ transparent cube named cuvette was cleaned by using acetone solution and dropper was used to drop the extract solution into the cube. The cuvette filled with extract solution was put onto the sample compartment and the analysis begun. The process was repeated three times to get the average reading.

2.5 Preparation of Substrate

The substrate used is SS316L which was a marine grade stainless steel. The specimen was obtained from a supplier in Terengganu and was cut into small rectangular shape with a dimension of 25 mm x 25 mm x 1 mm. Then, polishing of the steel is done using polished machine with abrasive paper grade 100, 120 and 180. The necessity of polishing the SS316L was to ensure the layer of carbon oxide is removed so that it will not interfere with the later procedure. Simple preservation method for stainless steel was carried out regularly until it is coated using sponge, distilled water and soap respectively.

2.6 Coating Formulation

Coating formulated involved several materials such as KTH 1K Alkyd Finish, general thinner and OLE. Simple mixing method is done according to the formulation in table 1. The mixture was stirred in a beaker on stir plate for 1 hour until it is homogenous. Finish product was sealed and stored in desiccator until used. The volume of extract solution was calculated based on the Equation 1:

$$\frac{\text{Volume of Solution (ml)}}{\text{Volume of Solvent (ml)}} \times 100 \quad (1)$$

Table 1. Coating Formulation.

Type of Coating	Volume of Paint (ml)	Extract (%)	Thinner (ml)
1 (control)	350	0	35
2	350	10	35
3	350	20	35

2.7 Immersion Test

Several beakers containing a fix amount of distilled water, deionized water and seawater was prepared. Different concentrations of OLE (0%, 10% and 20%) coating are used to coat the SS316L and later the substrate are placed inside the beaker hanging. Experiment is conducted for a period of 30 days and in each subsequent 10, 20 and 30 days, substrates of different concentration were taken out for analysis. The immersion test was conducted in an enclosed environment to avoid any disturbance.

3. Results and Discussion

3.1 Functional Group of Olive Leaves

Fourier Transform Infrared (FTIR) spectroscopy results in figure 1 show several peaks indicating the presence of active compounds in OLE. The compound identified is an important aspect for further understanding on the inhibiting property of the extract. Through the study of compounds identified will pose strong clarification on the mechanism of OLE application as green inhibitor. Base on figure 1, OLE includes major components such as oleuropein and hydroxytyrosol. The primary components of compound can be identified from the structures of phenols O-H, carboxylic acid C=O and alkenes C=C stretching nodes which are observed at 3329 cm^{-1} and 1733 cm^{-1} . The band at 1112 cm^{-1} could be assigned to the C-OH vibrations of the protein of olive leaves or C-N stretching vibration of the amine. The characteristic peak at 2927 cm^{-1} may be attributed to the C-H stretching node while the peak of 1651 cm^{-1} corresponds to the presence of amide I. Finally, the peak observed at 1040 cm^{-1} could be the indicator of C-N stretching vibration of the amine group. Table 2 shows the major functional groups found in OLE.

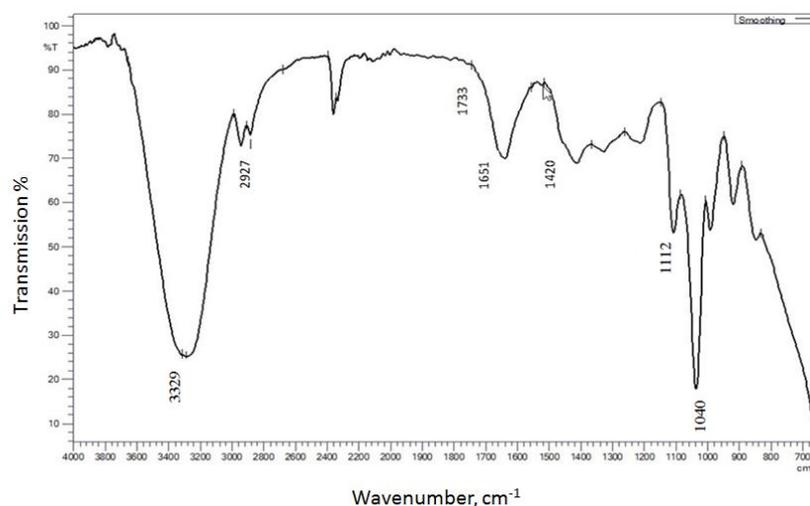
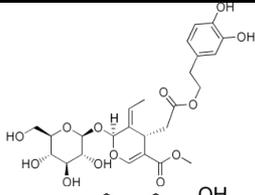
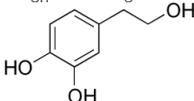
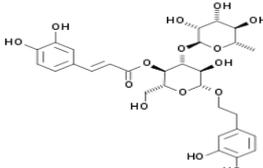
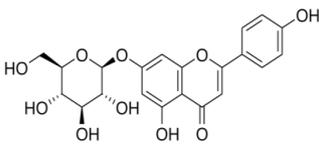


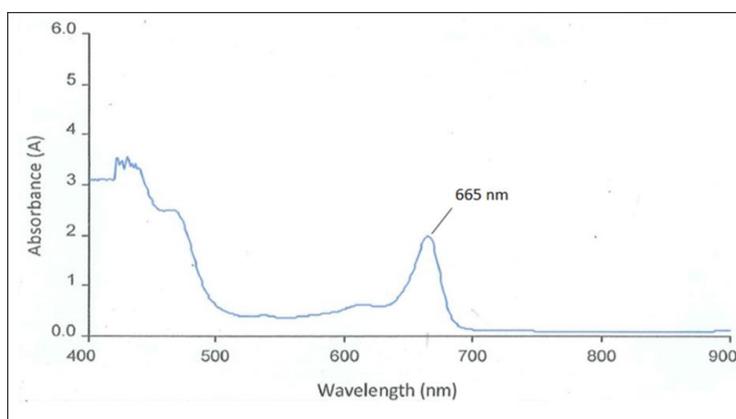
Figure 1. FTIR spectrum for olive leaf.

Table 2. Chemical Structures of the main Phenolic Compound in the Leaf of Olive [20].

Phenolic Compound	Chemical Formula	Chemical Class
Oleuropein		Secoiridoids
Hydroxytyrosol		Phenolic alcohol
Verbascoside		Flavonoids
Apigenin-7-glucoside		Flavonoids

3.2 Absorption Spectrum of Olive Leaves Extract

Absorbance is a measurable quantity that possesses a linear characteristic with the analyte concentration in comparison to the large range of concentration. By using UV-Vis spectroscopy, a measurement on the amount of light absorbed at a particular wavelength as the light passes through a sample or substances are recorded. Figure 2 shows the absorption spectra of diluted OLE. UV-spectrum was collected from 400 to 900 nm at 200 nm/min. The analysis was carried out using the lambda 25 software on the computer which connected to the equipment via cables. The absorption for the extract is 665 nm; respectively.

**Figure 2.** UV-Vis spectrum of OLE.

3.3 Electrochemical Impedance Spectroscopy (EIS)

Nyquist plots is used as the presentation of data collected for assessing the stability of the system. The real impedance (Z') was indicated at x-axis while the imaginary impedance (Z'') was indicated at y-axis. The parameter showed included the difference of OLE concentration use in the coating which was 0%, 10% and 20%. Base on the Nyquist plot in figure 3, capacitive semi-circle were shown by the curves obtained. Since there is no change in the shape of the curves with the difference of OLE concentration, it indicates that corrosion mechanism is maintained throughout the addition of inhibitor. The value the charge transfer resistance of the stainless steel as shown in table 3 significantly increased as the presence of OLE increased. However, the potential resistance (R_p) of 20% OLE at room temperature is lower than 10% OLE. The decrease of the R_p occurred due to the limitation in concentration. Increasing R_p leads to an increase of inhibitor efficiency. It can be conclude from the result that the ratio of OLE in the coating is found limited to 10% due to further increase of extract cause the potential resistance to reduce. Thus, increase in the concentration of extract beyond its optimum limit will results in inhomogeneity of coating sample [21].

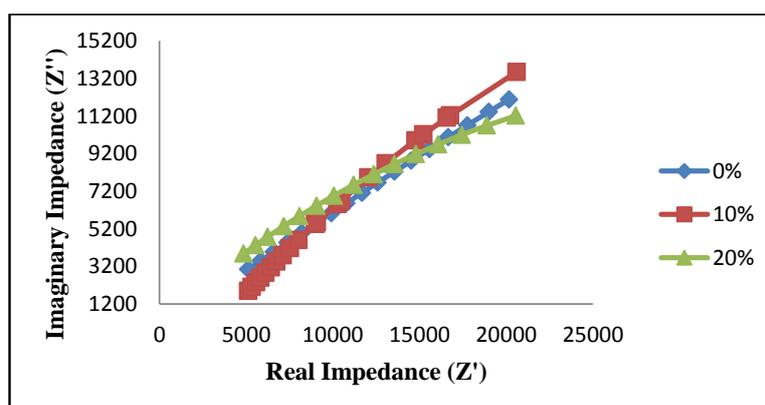


Figure 3. Nyquist plot for 30 Days in Seawater at room temperature.

Table 3. Impedance Parameter of SS316L in seawater for 30 days immersion.

Type of Coating	$R_p(\Omega)$
OLE 0%	75949
OLE 10%	105040
OLE 20%	75381

3.4 Potentiodynamic Polarization

Tafel plot is employed for the presentation of polarization measurement. From figure 4 and figure 5, the value of corrosion potential (E_{corr}) is seen to shift more towards the positive direction as the extract of olive leaf is introduced. The E_{corr} value of the samples in seawater and distilled water shown in table 4 and table 5 increases as there is presence of OLE which is -0.26573 V/SCE to -0.18793 V/SCE and -0.10672 to -0.06135 V/SCE respectively. Since the E_{corr} shift toward more positive direction, it elucidates that OLE act as a mixed-type inhibitor.

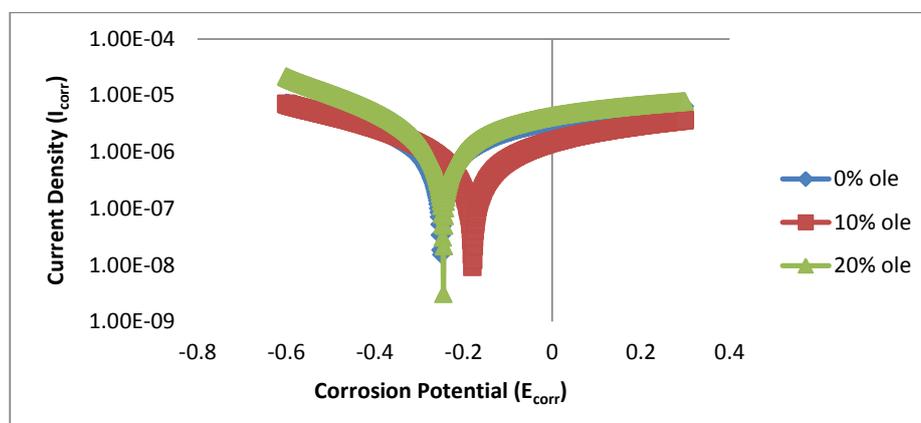


Figure 4. Potentiodynamic polarization of SS316L for 30 days in the medium of seawater.

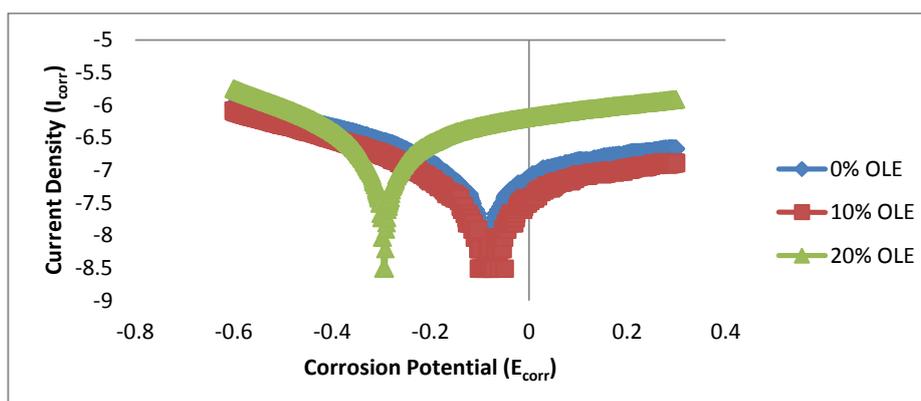


Figure 5. Potentiodynamic polarization of SS316L for 30 days in the medium of distilled water.

However, the value of E_{corr} drop to -0.25489 V/SCE and -0.29449 V/SCE for both samples in seawater and distilled water as the OLE concentration increases to 20%. As the value of corrosion potential increase the corrosion rate will decrease. Thus, increase in the concentration of OLE will results in higher corrosion potential. The low value of corrosion rate is due to the thin layer formed on steel substrate to protect the steel from corrosion [22]. The studied done previously on the inhibitor used was affected both in anodic and cathodic sites in Tafel plot with E_{corr} value towards more noble values and this phenomenon can be used to derived that inhibitor act as a mixed-type inhibitor [23].

Table 4. The electrochemical parameter of SS316L in seawater for 30 days immersion.

Type of Coating	E_{corr} (V/SCE)	I_{corr} (A.cm ²)	Corrosion rate (mm/year)
Olive Leaves Extract 0%	-0.26573	6.88E-06	0.079930
Olive Leaves Extract 10%	-0.18793	1.68E-06	0.019463
Olive Leaves Extract 20%	-0.25489	3.95E-06	0.045848

Table 5. The electrochemical parameter of SS316L in distilled water for 30 days immersion.

Type of Coating	E_{corr} (V/SCE)	I_{corr} (A.cm ²)	Corrosion rate (mm/year)
Olive Leaves Extract 0%	-0.10672	5.68E-07	0.006602
Olive Leaves Extract 10%	-0.06135	1.33E-07	0.001542
Olive Leaves Extract 20%	-0.29449	3.46E-07	0.004019

Table 4, Table 5 and Table 6 provide an elucidation that corrosion rate decrease as the value of current density (I_{corr}) becomes lower due to the presence of OLE. 10% of OLE has lower corrosion rate than 20% of OLE in both seawater and distilled water due to the ratio of the extract is proved to be limited at 10% in the study. According to another studies, the inhibition of steel increases with the increase of an organic extract concentration [24]. Moreover, through the samples immersed in seawater shows decrease in current density from 6.88×10^{-6} A.cm² to 1.68×10^{-6} A.cm². This behavior proved that OLE acts as a good corrosion inhibitor when integrated in paint for immersed stainless steel in marine medium.

Table 6. The electrochemical parameter of SS316L in deionized water for 30 days immersion.

Type of Coating	E_{corr} (V/SCE)	I_{corr} (A.cm ²)	Corrosion rate (mm/year)
Olive Leaves Extract 0%	-0.268934	4.13E-08	0.000480385
Olive Leaves Extract 10%	0.273840	3.53E-08	0.000410365
Olive Leaves Extract 20%	0.097405	4.21E-08	0.000489658

The potentiodynamic polarization curve obtained from the analysis of the samples immersed for 30 days in deionized water shows almost a similar rate of corrosion rate. Deionized water or demineralized water is ultrapure water usually used in microelectronics and instrument manufacture. Although most people hold the opinion that deionized water is a very corrosive medium, a study by National Aeronautics and Space Administration (NASA) shows the medium have no such effect on common metals such as aluminum and stainless steel [25].

3.5 Scanning Electron Microscope

The primary analysis was subjected to the coated surface of SS316L which has been immersed in seawater medium after 30 days; respectively. Both figure 6(a) and figure 6(b) show a similar result of the presence of pores or pin holes. Pore or pin holes are basically tiny spots indicating the absence of coated layer and varied in its dimension. There are several typical causes of this phenomenon such as the rupture of liquid coating caused by contamination, picking of coating by roll and non-uniform flow of coating when it is applied. Other related sources might be due incompatibility of the coating with the surface of the substrate. However, this theory is rejected since the selection of the components in the coating is selected based on the previous study conducted in several researches which used the same type of constituents and differs only in the type of extract use. Thus, it indicates that the incompatibility of the coating with the surface is not the factor that caused the surface to possess this state. Through the researcher observation, there are two possible causes of the phenomenon which are

irregularities on metal and non-uniform flow of coating when it is applied. The evidence on the first suggestion are supported in figure 6(d) that showed the surface of a naked SS316L possessing several dark spots indicating the irregularities or roughness of the surface. The second causes occurred due to the unsuitable method of applying the coating onto the SS316L surface using a simple paint brush in several directions.

Generally, porosity is identified as an undesirable trait since it will expose the substrate to corrosive agents, deleteriously influence density, electrical property and diffusion characteristic as well as reduce mechanical properties, [26]. Various tiny pores are observed in figure 6(a) and figure 6(b) with dimension ranging from 10 μm to 40 μm . Figure 6(c) shows the SEM micrograph of the coated surface of SS316L with 10% OLE, it differ compare to previous two result in which the surface shows a muddy layer with cracks in several places. Cracks can be considered as another form of pores which extend usually in a direction parallel to the surface. Type of cracks includes transverse crack and enclosed crack. Probable causes of the phenomenon are due to the application of the coating was too thick, coating flash too quickly or at too high temperature. It may occur in an irregular pattern where the coating stresses are rather uniform. It may be linear, following brush marks occur in a curved pattern with the crack meeting and intersecting in a rather typical form. The problem can be overcome easily by using a proper resins and pigments with the addition of material specifically for reducing interior stresses within the coating. Through the observation, a few cracks can be seen on several places especially on the area where coating appeared thicker.

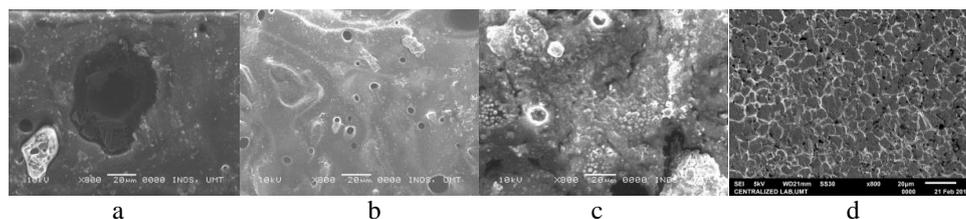


Figure 6. SEM micrographs of SS316L surface (a) after 30 days of immersion with 0% OLE coating (b) after 30 days of immersion with 20% OLE coating (c) after 30 days of immersion with 10% OLE coating (d) naked.

4. Conclusion

In summary, inhibitor plays a significant role in endowing protection against corrosion. The olive leaves extraction was successfully prepared through a maceration method with an organic solvent. The absorption spectra by UV-Vis spectroscopy showed the absorption for the extract is 665 nm; respectively. Through the analysis of FTIR spectroscopy several functional groups which indicate the presence of antioxidant compound such as oleuropein and hydroxytyrosol was identified. The extract was applied as an additive providing the anti-corrosion property with different concentration (0%, 10% & 20%) for the means of investigating the efficiency of OLE as corrosion inhibitor. Immersion test for a month was carried out in the laboratory using three different media which were seawater, distilled water and deionized water. By the end of 30 days, sample was taken out and analyzed with EIS spectroscopy, PP and SEM for a detailed corrosion rate determination. EIS measurement indicates that the inhibitor of corrosion on SS316L is due to the protective film on metal surface and inhibition increases as the extract concentration increases. As the presence of OLE increases, the values of potential resistance (R_p) increase accordingly. However, ratio of OLE is found limited to only 10%. Further increase in the concentration of extract would cause inhomogeneity in coating paint sample. The pitting potential of SS316L shifted to more positive values with the addition of extract indicating resistance to pitting corrosion increase. The low value of corrosion potential

(E_{corr}) indicates higher corrosion rate. The value of corrosion current density (I_{corr}) increases with absence of OLE. Base on the results from all the samples tested, the substrate with 10% OLE coating showed the highest inhibition rate of corrosion followed by 20% and 0% OLE coating. In a conclusion, OLE possesses a good potential as an excellent inhibitor for stainless steel.

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