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Corrosion Behaviour of Aluminium and AA5754 Alloy in 1% Acetic Acid Solution in Presence of Laurel Oil

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Electrochemical investigations show that aluminium and AA5754 alloy have certain corrosion activity in 1% acetic acid solution. Results indicate that aluminium has better corrosion resistance to the action of 1% acetic acid than its AA5754 alloy. The addition of ethanol solution of laurel oil in acetic acid decreases the values of corrosion current densities and increases the polarization resistance. Corrosion inhibition effect of laurel oil is confirmed by SEM examination.

Keywords: aluminium, aluminium alloy, acetic acid, laurel oil, corrosion inhibition.

1. INTRODUCTION

Aluminium is widely used in a plenty of industrial applications such as constructions, electrical engineering, transport, and especially in food industry for the manufacture of processing, production, storage and transportation equipment and machinery. Despite the good properties of aluminium, aluminium and its alloys are not perfect materials for engineering applications in all environment, since they suffer from corrosion caused by chemical interactions with their surroundings. High corrosivity environment for aluminium in food industry are foodstuffs with pH 3 – 5, such as fruit juices, jams and acidic canned fruits or hot gravies, sauces as well as dressings, vegetables and fish pickled in brines with 1 –3 % salt [1]. In particular, acetic acid needs to be taken into corrosion consideration, due to its wide usage in food industry (vegetable and fish pickling), and its representative properties among acids and juices in fruits, vegetables and other organic materials that can corrode metals. Although aluminium has a good resistance to acetic acid solution at room temperature, aluminium can corrode in almost any concentration of acetic acid at any temperature if the acid is contaminated with the proper species [2]. It has been established that cooking of acidic and low acidic foods in aluminium saucepans or foil causes leaching of the metal into food products [3-5]. It was reported in some publications that foods which were manufactured and stored in aluminium cans, showed an increased aluminium content [6]. Such metal release can, eventually, cause health risks for consumers [7]. One of the most common solution for reducing corrosion process is the addition of organic inhibitors into aggressive corrosion medium. Studying plants from Mediterranean region as corrosion inhibitors, we have shown that essential oils of lavender, laurel leaves, dill and basil have a very good inhibition properties preventing aluminium corrosion in acidic and neutral solutions [8-11].

Since a few papers deal with some aspects of aluminium and its alloys behaviour in contact with acetic acid solution [12,13], we assume that it is necessary to investigate the aluminium and AA5754 alloy behaviour in 1% acetic acid solution in absence and presence of *Laurus nobilis* L. essential oil (laurel oil). This study was conducted by using potentiodynamic polarization and linear polarization methods, while the aluminium and AA5754 alloy surfaces were examined by scanning electron microscopy (SEM).

2. EXPERIMENTAL

The chemical composition (wt %) of pure aluminium used in this study is Fe 0.08, Si 0.06, Cu 0.001 and balanced Al (99.85); while the composition of the AA5754 alloy sample is Mg 3.10, Si 0.40, Fe 0.40, Cu 0.10, Mn 0.50, Cr 0.30, Zn 0.20, Ti 0.15 and balanced Al (94.85).

The basic solution was 1% acetic acid (HAc). The initial solution of inhibitor was prepared by addition of laurel oil as 30% ethanol solution into basic solution (1% HAc), and was used in concentration range from 10 to 50 ppm. The inhibitor concentration were calculated and expressed in manuscript as mg L^{-1}, because the density of laurel oil is known. The chemical composition and density of laurel oil were reported previously [10]. Potentiodynamic polarization and linear polarization measurements were carried out in a three electrode glass cell containing 500 mL of electrolyte. A saturated calomel electrode (SCE) was used as reference and two graphite electrodes were used as auxiliary electrodes. The working electrode (aluminium or AA5754 alloy) was mounted in Teflon holder where 1 cm^2 of surface was exposed to aggressive solution. The sample surface was abraded with emery paper to a 1200 metallographic finish and rinsed with distilled water, before each measurements and each experiment was done in triplicate for ensuring the reproducibility. The electrochemical measurements were performed in the test solution after reaching the open-circuit potential (E_{ocp}). Potentiodynamic polarization studies were performed at scan rate of 0.5 mV s^{-1} in the potential range from ±150 mV with respect to the E_{ocp}, and the all potentials reported refer to SCE. All the electrochemical experiments were conducted using a computer-controlled with a Potentiostat type VersaSTAT 3 (Princeton Applied Research).

Linear polarization measurements were performed in in the potential range ±20 mV from the corrosion potential with scanning rate of 0.16 mV s^{-1}. Since the polarization curves obtained are not completely linear in this potential domain, the polarization resistance, R_{p}, was determined from the data collected in potential range ±5 mV from the corrosion potential.

The surface morphology of the aluminium and AA5754 alloy samples, after 24 h immersion in 1% acetic acid solution in absence and presence of 45 mg L^{-1} of laurel oil was recorded on JEOL JSM-6460 scanning electron microscope. The aluminium and AA5754 alloy coupons were prepared as described previously for electrochemical measuring.

3. RESULTS AND DISCUSSION

3.1. Polarization measurements

Although aluminium has a good resistance to almost all the concentrations of acetic acid at room temperature, care must be taken that the metal is free of other impurities such as iron, copper, tin and lead even in traces [2]. With increasing purity of aluminium, its resistance to acetic acid solution increases, and 99.5% aluminium can be used for the majority of engineering purposes, but components added to its alloy can increase the corrosion of aluminium in acetic acid solution [14]. Since the metal corrosion occurs via electrochemical reactions at the interface between the metal and an electrolyte solution, electrochemical techniques are ideal for the study of corrosion processes.

Results of potentiodynamic polarization measurements obtained for aluminium and AA5754 alloy in 1% acetic acid solution are represented in Figures 1a and 1b., respectively. It can be seen from these figures that the both metals investigated, aluminium and its alloy show certain corrosion activity in 1% acetic acid solution. From polarization curves it is evident that the dissolution of metal is uniform, what is not an unexpected behaviour for aluminium in acidic solution (pH value of 1% acetic acid in this investigation is 3.1). It is well known that aluminium resistance is related to the thin and compact layer of naturally formed oxide on aluminium surface, but this oxide layer is stable only in pH range 4-8. Lower or higher pH values caused prominent destroying of protective layer and thus the significant metal dissolution [15].

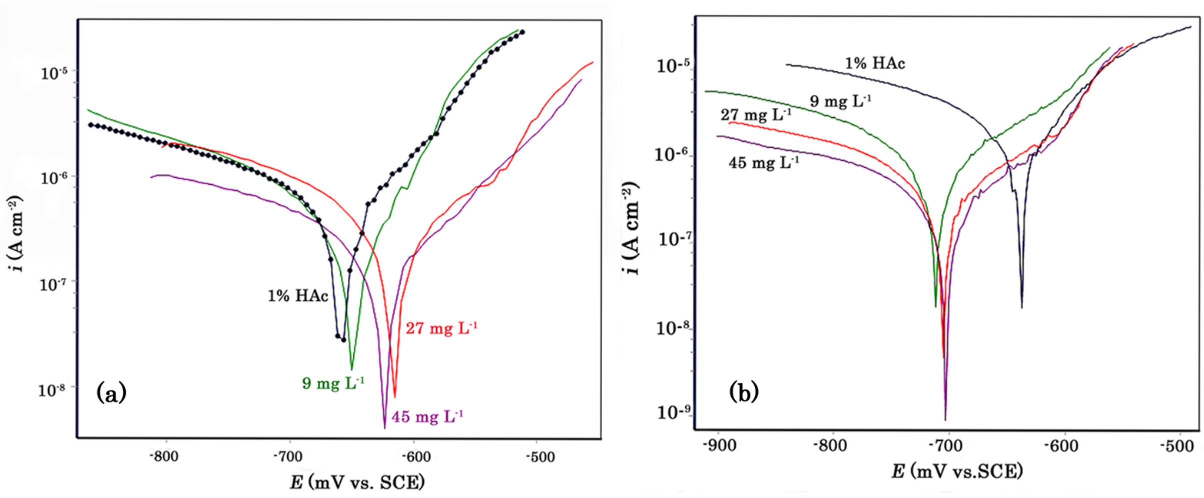


Figure 1. Potentiodynamic polarization curves obtained for aluminium (a) and AA5754 alloy (b) in 1% acetic acid solution in the absence and presence of laurel oil at 298 K.

Inspection of Figures 1a and 1b also showed that the addition of laurel oil causes a remarkable decrease in current density values for aluminium as well as for the alloy. However it must be noted that laurel oil has different effect on corrosion behaviour of aluminium in relation to the aluminium alloy. According to the corrosion potential values E_{corr}, being in the case of AA5754 alloy more negative with increasing concentration of laurel oil, the presence of laurel oil in acetic acid caused corrosion potentials shifting to more noble values for pure aluminium, with reference to the blank solution. As can be seen, laurel oil has a greater influence on anodic reaction on aluminium, i.e. metal dissolution reaction is more inhibited, while in the case of alloy, cathodic reaction of hydrogen evolution is more suppressed. Parameters obtained from potentiodynamic polarization curves such as corrosion current densities (i_{corr}), corrosion potential (E_{corr}), cathodic Tafel slope (b_c), anodic Tafel slope (b_c) and inhibition efficiency (η) are listed in Table 1.

Table 1. Potentiodynamic polarization parameters obtained for aluminium and aluminium alloy in 1% acetic acid solution in absence and presence of different concentrations of laurel oil at 298 K.

Material	Inhibitor conc. (mg L^{-1})	E_{corr} (mV_{SCE})	i_{corr} (µA cm^{-2})	$-b_c$ (mV dec^{-1})	$-b_a$ (mV dec^{-1})	η (%)
Al	0	-664	6.032	175	63	-
	9	-648	4.784	172	69	20.7
	18	-650	4.022	162	73	33.3
	27	-615	3.165	165	70	47.5
	36	-617	2.458	168	70	59.3
	45	-621	1.873	163	73	68.9
AA5754 alloy	0	-640	17.033	168	67	-
	9	-712	9.081	156	75	46.6
	18	-703	6.225	140	76	63.4
	27	-706	4.312	139	78	74.7
	36	-674	3.139	142	77	81.6
	45	-702	2.659	147	74	84.4

According to the corrosion current density values given in Table 1., it is evident that aluminium is more corrosion resistant in 1% acetic acid than AA5754 alloy (6.032 µA cm^{-2} and 17.033 µA cm^{-2}, respectively). Presumably, this can be attributed to the fraction of impurities present in this aluminium alloy, especially iron (0.4%) and copper (0.1%). Even in the case when their content in aluminium is low, these second-phase impurity precipitates simply degrade the corrosion resistance of Al alloy by increasing the cathodic volume fraction [16]. Moreover, the alloying elements are often distributed not only in the solid solution of aluminium, but also in intermetallic particles, which play a crucial role in the corrosion behavior of Al alloys. By increasing the amount of Mg content in aluminium alloy, the strength of the alloy is significantly increased and improved [17].

On the other hand, with higher Mg content in aluminium alloy, the so-called α-phase (Al_3Mg_2) content in the alloy increases, and usually precipitates at the grain boundaries or on the alloy surface,and corrosion attack can takes place near these particles [18].

From the Table 1, it is clear that the addition of laurel oil decreases the corrosion rate of pure aluminium as well as AA5754 alloy. The values of b_c and b_a do not change significantly, indicating that laurel oil addition to acetic acid solution does not change the mechanism of aluminium dissolution and hydrogen evolution reaction. It means that components present in laurel oil (such as 1,8-cineole) were adsorbed on aluminium and AA5754 alloy surface by simply blocking the active sites.

The inhibition efficiency was evaluated from the Eq (1):

$$\eta_p(\%) = \left(\frac{i_{corr} - i^0_{corr}}{i_{corr}} \right) \times 100 \qquad (1)$$

where i_{corr} and i^0_{corr} are the corrosion current densities in the absence and presence of inhibitor.

The calculated values of inhibitor efficiency η_p increases with increasing concentration of laurel oil and are higher for AA5754 alloy corrosion in 1% HAc solution than those obtained for pure aluminium at the same concentration of the oil added. Therefore, the maximum inhibition efficiency of 45 mg L^{-1} laurel oil added was 68.9% on aluminium and 84.4% on AA5754 alloy, respectively.

Typically linear polarization plots in an arrow potential range obtained for aluminium and AA5754 alloy in 1% acetic acid solution, and acetic acid solution with 45 mg L^{-1} laurel oil solution added are shown in Figure 2.

Figure 2. shows that in the case when laurel oil was added the slope of polariazation curves decreases, indicating that polarization resistance increase. The polarization resistance, R_p, corrosion potential, E_{corr}, and corrosion current density (i_{corr}) are determined by a linear regression calculation at current density–potential curve close to the corrosion potential and are given in Table 2.

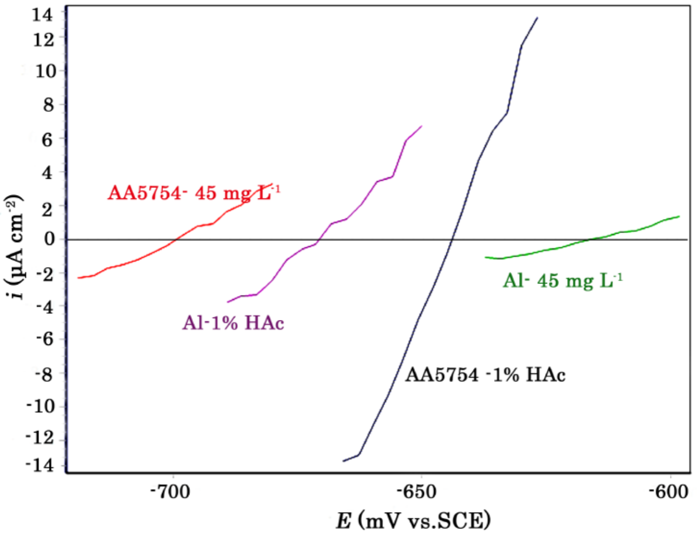


Figure 2. Linear polarization curves for aluminium and AA5754 alloy in 1% acetic acid solution in the absence and presence of 45 mg L^{-1} laurel oil at 298 K.

Table 2. Corrosion parameters of aluminium and aluminium alloy in 1% acetic acid solution in absence and presence of different concentrations of laurel oil determined by the linear polarization method at 298 K.

Material	Inhibitor conc. (mg L^{-1})	E_{corr} (mV_{SCE})	R_p (kΩ cm^2)	i_{corr} (μA cm^{-2})	B (mV)	η (%)
Al	0	-668	3.44	5.838	20.1	-
	9	-645	4.60	4.649	21.4	25.2
	18	-653	5.84	3.735	21.8	41.0
	27	-611	8.73	2.441	21.3	60.5
	36	-616	10.20	2.107	21.5	66.2
	45	-625	14.95	1.464	21.9	76.8
AA5754 alloy	0	-644	1.44	14.442	20.8	-
	9	-715	3.61	6.053	21.9	60.1
	18	-702	4.59	4.637	21.3	68.6
	27	-703	6.13	3.540	21.7	76.5
	36	-672	6.71	3.221	21.6	78.5
	45	-701	7.49	2.857	21.4	80.7

Using the experimentally determined values of polarization resistance R_p, corrosion current densities can be calculated according to Stern–Geary equation (2):

$$i_{corr} = \frac{b_a \cdot b_c}{2.303(b_a + b_c)R_p} = \frac{B}{R_p} \quad (2)$$

where b_c and b_a are cathodic and anodic Tafel slope, respectively; R_p is polarization resistance, and B is Stern-Geary coefficient [19].

The data given in Table 2. show that pure aluminium is more resistant than its alloy since the R_p value for aluminium is higher (3.44 kΩ cm^2) than that obtained for AA5754 alloy (1.44 kΩ cm^2). This observation confirms that these results are in good agreement with those obtained from Tafel extrapolation method. The values of R_p constantly increase with increasing laurel oil concentration, while the current density values decrease. The polarization resistance value is the highest at concentration of 45 mgL^{-1} laurel oil, while higher R_p value indicates lower corrosion rate. It must be noted that Stern-Geary coefficients, B were not changed significantly after addition of laurel oil to acetic acid solution (about 20 mV without inhibitor and about 22 mV with inhibitor), being typical for the metallic corrosion at active state. Figure 2 shows that in presence of laurel oil, the corrosion potential of aluminium shifts towards more noble values, while for alloy this shift of corrosion potential is opposite, and shifted to more negative direction. Therefore, in agreement with the Tafel extrapolation method, laurel oil is an anodic corrosion inhibitor for aluminium and cathodic corrosion inhibitor for AA5754 alloy.

Since the electrochemical theory assumes that $1/R_p$ is directly proportional to the corrosion rate, the surface coverage and inhibition efficiency, η (%) of laurel oil was calculated from (R_p) values obtained from linear polarization data using Eq (3):

$$\eta_{LP}(\%) = \left(\frac{R_p^0 - R_p}{R_p^0} \right) \times 100 \qquad (3)$$

where R_p^0 and R_p are polarization resistance with and without inhibitor, respectively.

Inhibition efficiency values calculated from potentiodynamic polarization and linear polarization methods are not the same but have the same order (with increasing concentration of laurel oil inhibition efficiency increases). According some authors [20] the full agreement of the data obtained by using Tafel extrapolation and linear polarization is practically impossible to obtain, because during the anodic and cathodic polarization the morphological structure and roughness of the metal surface changes. Moreover, the polarization in wide potential range does not interfere on corrosion system, since the corrosion potentials, E_{corr} values of aluminum and AA5754 alloy, obtained from linear polarization, are very close to those obtained by potentiodynamic polarization (they differ only by ± 4 mV from E_{corr} specified in a wider potential area).

3.2. Adsorption isotherms

To ascertain the nature of adsorption, the surface coverage values for laurel oil obtained from potentiodynamic polarization measurements were fitted into different adsorption isotherm models and correlation coefficients (R^2) were used to determine the best fit which was obtained with Freundlich isotherm (Fig. 3a) for aluminium and Temkin isotherm model for AA5754 alloy (Fig.3b).

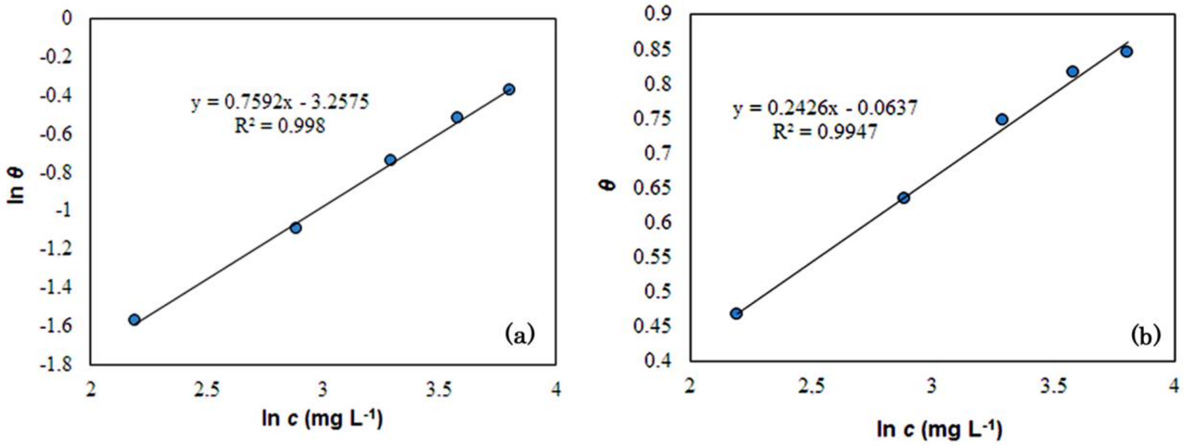


Figure 3. Freundlich isotherm (a) and Temkin isotherm (b) plots for the adsorption of laurel oil on aluminium and AA5754 alloy surface, respectively at 298 K.

It is found that the adsorption behaviour of laurel oil on aluminium can very well be described by the Freundlich adsorption isotherm since the correlation coefficient is 0.998.

Freundlich adsorption isotherm is given by the equations (6) and (7):

$$\theta = K \cdot c^{1/n} \qquad (6)$$

$$\ln\theta = \ln K + \frac{1}{n}\ln c \qquad (7)$$

where K is a constant indicated a relative adsorption capacity of the adsorbent (inhibitor molecules) and n is a constant indicated an intensity of adsorption [21]. The Freundlich expression is an exponential equation and therefore assumes that as the adsorbate (inhibitor) concentration increases, the concentration of inhibitor on the metal surface increases, too.

Temkin isotherm characterizes chemisorption of uncharged molecules on a heterogeneous surface [22] and is given by equation (8):

$$e^{f\cdot\theta} = K\cdot c \qquad (8)$$

where K is the adsorption equilibrium constant and f the molecular interaction constant - a coefficient expressing the interaction between adsorbed and adsorbing molecules.

$$\theta = \left(\frac{1}{f}\right)\ln K + \left(\frac{1}{f}\right)\ln c \quad (9)$$

Eq. (9) is the linear form of the Temkin isotherm, thus, the plot θ vs. ln c can give a straight line with a slope of (1/f) and an intercept of [(1/f) ln K], which is represent in Fig.3b. The applicability of the Temkin adsorption isotherm verifies the assumption of monolayer adsorption on an energetically uniform heterogeneous electrode surface, with interaction in the adsorption layer [22]. Temkin isotherm favours the assumption of a chemisorptive bond between metal and inhibitor molecules i.e. the unshared electron pairs in oxygen atom of 1,8-cineole could interact with p-orbital of Al to provide a protective chemisorbed film. Due to the fact that laurel oil is a mixture of various compounds containing oxygen and double bonds it is difficult to decide which of these components are responsible for this inhibition. Taking into account that the chemical composition of *Laurus nobilis* L. oil [10] reveals that 1,8-cineole is the major compound (46%), it can be assumed that this compound is largely responsible for the corrosion inhibition. Moreover, various adsorption active centres (oxygen atoms, double bonds etc.) are able to act synergistically to forming polymeric complex.

K_{ads} value can be related to the free energy of adsorption (ΔG^0_{ads}) by the following equation (10):

$$\Delta G^0_{ads} = -RT\ln(K\times c_{H_2O}) \qquad (10)$$

where c_{H_2O} is water concentration in solution expressed in mg L^{-1}, R (8.314 J mol^{-1} K^{-1}) is the universal gas constant and T (K) is temperature. It must be emphasized that the concentration unit of water molecules has to be similar to that of inhibitor and therefore the unit of c_{H_2O} is given in mg L^{-1} with the value of approximate 1 $\times 10^6$.

The adsorption parameters obtained from the given adsorption isotherms are listed in Table 3.

Table 3. Laurel oil adsorption parameters on aluminium and alloy in 1% HAc solution obtained from potentiodynamic polarization measurement at 298 K.

Materials	*Isotherm*	*1/n*	*f*	K (L/mg)	$-\Delta G^0$ (kJ/mol)
Al	Freundlich	0.759	-	0.038	20.45
AA5754 alloy	Temkin	-	4.122	0.769	33.57

Results indicate that adsorption equilibrium constant (*K*) value is higher for adsorption of laurel oil on AA5754 alloy (0.769 L mg^{-1}) than for adsorption on aluminium surface (0.038 L mg^{-1}). Larger value of *K* means better inhibition efficiency of inhibitor and this conclusion is in agreement with previously presented results. Moreover, the calculated free energy of adsorption (ΔG^0_{ads}) obtained for laurel oil adsorption on AA5754 alloy is -33.57 kJ mol^{-1} suggesting the mixed type adsorption (physisorption and chemisorption), while the value of -20.45 kJ mol^{-1} obtained for adsorption of inhibitor molecules on aluminium is consistent with the physisorption mechanism [23].

Different adsorption behavior of laurel oil on aluminium alloy from adsorption behavior on aluminium surface, can be due to the surface inhomogeneity. It must be noted that when aluminium is introduced into acid, acid first dissolves the air-formed film on aluminium, and then the rapid attack on the bare metal start. On the other hand, the reduction of oxygen present in solution and low level of hydrogen evolution can together increase pH value around intermetalic particles and intensifies dissolution of the oxide film on these particles, and accelerating acid attack on AA5754 alloy.

It can be concluded that adsorption of laurel oil molecules on alloy surface occurs first of all at that defective spots (cathodic intermetalic particles) were oxide layer is dissolved [24].

3.3. SEM analysis

The aluminium and aluminium alloy samples were immersed in 1 % HAc solution for 24 h, without and with addition of 45 mg L^{-1} of laurel oil solution and their surface morphology was examined by scaning electron microscopy. The SEM micrographs recorded are shown in Figs. 4 and 5.

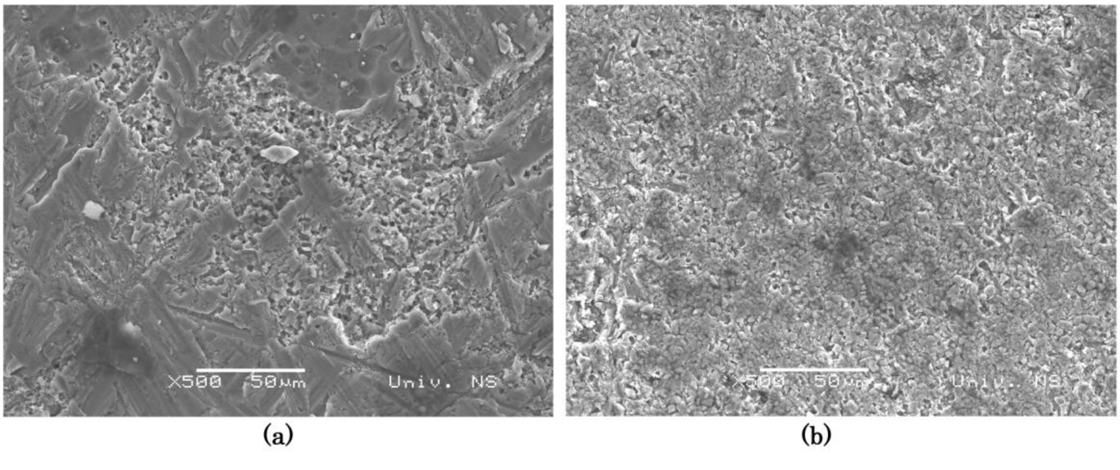
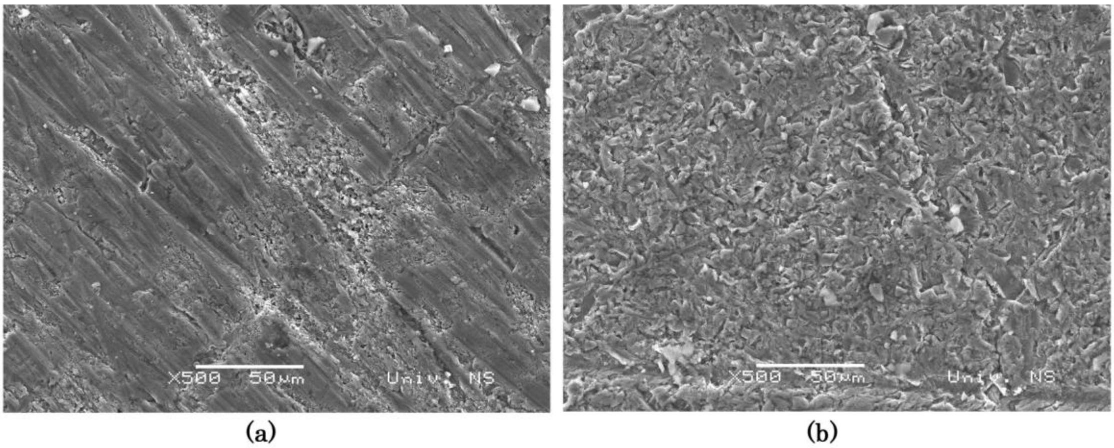


Figure 4. Scanning electron micrographs of the AA5754 alloy surface after 24 h immersed at 25°C in 1% acetic acid solution (a) and in 1% acetic acid solution with 45 mg L^{-1} laurel oil.

The SEM micrographs (Fig. 4) clearly show that the surface is quite damaged in the absence of the inhibitor, and a severe dissolution of the alloy surface in contact with acetic acid solution was noticed. This obviously resulted in an increase in surface porosity together with small holes formation on metal surface (Fig. 4a). In the presence of laurel oil as can be seen on Fig. 4b the rate of corrosion is suppressed, and there exist only small acid corrosion cracks on the aluminium surface. Close observation of SEM image indicates the formation of some sort of layer on the AA5754 alloy surface.

The SEM image of corroded aluminium surface is given in Fig. 5a showing degradation of surface, with more or less uniform attack in acetic acid. By comparison of the damaged size on aluminum and its alloys, we noticed that the cracks on the aluminium are considerably smaller than that observed on its alloy surface. Fig. 5b gives again evidence that addition of laurel oil to acetic acid solution causes a formation of thick and protective layer of adsorbed inhibitor on aluminium surface.



(a) (b)

Figure 5. Scanning electron micrographs of the aluminium surface after 24 h immersed at 25°C in 1% acetic acid solution (a) and in 1% acetic acid solution with 45 mg L^{-1} laurel oil.

4. CONCLUSIONS

The results obtained by experiments in this work clearly indicate that aluminium has better corrosion resistance to the action of 1% acetic acid solution than its AA5754 alloy. Electrochemical studies showed that addition of ethanol solution of laurel oil in acetic acid decreases the values of corrosion current densities and increases the polarization resistance. The maximum inhibition efficiency of 45 mg L^{-1} laurel oil added was 68.9% on aluminium and 84.4% on AA5754 alloy. The corrosion inhibition of laurel oil could be interpreted by adsorption of active molecule according to Freundlich isotherm on the pure aluminium surface and Temkin isotherm on AA5754 alloy surface. Laurel oil is an anodic corrosion inhibitor for aluminium and cathodic corrosion inhibitor for AA5754 alloy. Corrosion inhibition can be explained by the formation of the protective layer on aluminium and alloy surfaces, which is revealed by SEM studies.

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