Influence of green tea extract on corrosion of different metals in 0.5 mol dm^{-3} NaCl solution

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ABSTRACT

The corrosion behavior of copper, aluminum and stainless steel AISI 316L in 0.5 mol dm^{-3} NaCl solution without and with the addition of an aqueous-ethanol extract of green tea leaves at a temperature of 25°C was investigated. Electrochemical parameters where determined by linear polarization measurements and potentiodynamic polarization method, and the surface of the metal samples were investigated using an optical microscope after the polarization measurements. Results have shown that green tea extract acts as a corrosion inhibitor for copper aluminum and stainless steel.

Indexing terms/Keywords

Corrosion; green tea; polarization; aluminium; copper; AISI 316L

Academic Discipline And Sub-Disciplines

Electrochemistry

SUBJECT CLASSIFICATION

Environment Science (Green corrosion inhibitor)

TYPE (METHOD/APPROACH)

Corrosion study

Council for Innovative Research

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INTRODUCTION

Metals, such as copper, aluminum, and its alloys along with stainless steels, are used in almost all spheres of human activity today. Thus, copper, due to the extremely high electrical and thermal conductivity and good mechanical properties, has great application in electronics, in the manufacture of electrical conductors, heat exchangers, boilers, food and chemical industries etc [1,2]. Aluminium is widely applied as construction materials in aircraft and automotive industry, metallurgy, mechanical engineering, chemical and food industry because it has low density, good mechanical strength, which improves by alloying and heat treatment, and high corrosion resistance [3]. Stainless steels, due to its high corrosion resistance and excellent mechanical properties are used in chemical and food industry, as construction materials in building industry and the automotive and shipbuilding industries [4]. In terms of exploitation, products made from these metals are exposed to a variety of chemical, physical and biological influences that may adversely affect these products reducing their utility. This process unintentional destruction of materials by chemical, physical and biological agents is called corrosion. Metallic materials from corrosion processes are protected in different ways and one of them is the application of corrosion inhibitors. However, a large number of commercial corrosion inhibitors are synthetic chemicals which may be expesive and hazardous to living creatures and environments, so in recent years efforts have been made to replace it with natural non-toxic substances. Many investigators turned their attention to plant extracts, which have become important for investigation as environmental acceptable, readily available and renewable source for wide range of potential corrosion inhibitors [5-10]. This was the motivation for this work in which we investigated the possibility of corrosion inhibition of pure Cu, Al and stainless steel AISI 316L using green tea extract in NaCl solution. Green tea has been selected for investigation because it is easily accessible, inexpensive and renewable source of various phenolic compounds that have proven positive effect on human health [11-14]. Also, certain phenolic compounds have already shown themselves to be good corrosion inhibitors for various metals and alloys [15-19].

There are only few reports on the corrosion behavior of metals in the presence of green tea extract in the literature. Most investigations have focused on inhibitive effect of green tea extract on corrosion of different metals and alloys in acidic solution. Lotto and associates investigated inhibitive effect of green tea extract on corrosion of mild steel and aluminium alloy in sulphuric and hydrochloric acid [20-23]. The obtained results showed effective corrosion inhibition of the green tea extract on the aluminium alloy and mild steel in acidic solutions. Nofrizal and associates [24] in their investigation of corrosion inhibition of mild steel in 1.0 M HCl solution were found that adsorption of green tea extract constituents followed Langmuir adsorption isotherm and the calculated Gibb's free energy values indicated the physisorption of inhibitor on mild steel surface. The investigation of Rustandi and associates is only paper in Scopus database which deal with the corrosion inhibition effect of green tea extract in NaCl solution [25]. In their investigation of flow induced corrosion they found that the addition of mixture of 1000 ppm piper betle and 4000 ppm green tea extracts with Reynold number ranging from 0 up to 30000 reduced the corrosion rates of steel significantly with its approximately 90 % inhibitor efficiencies achieved.

1. EXPERIMENTAL

1.1. Materials and methods

The corrosion investigations were performed on different metal samples made form 99.99% Cu, 99.999% Al and AISI 316L which composition is showed in table 1:

Table 1. Chemical composition of AISI 316L

element	C	Si	Mn	P	S	Cr	Mo	Ni	Fe
w / %	0.02	1.00	2.00	0.045	0.030	17.00	2.00	10.00	rest

The lateral parts of working electrodes were insulated with Polyrepar resin so the only one side of metal samples was left uninsulated as electrode working area. The exposed electrode surface was abraded with different grades of emery papers (from 240 to 1500), degreased ultrasonically with ethanol and rinsed with deionized water. Electrochemical experiments were conducted in a three-electrode cell using a potentiostat/galvanostat (PAR Model 273A). A platinum sheet was used as counter electrode and a saturated calomel electrode (SCE) equipped with a Luggin capillary as a reference electrode.

The main electrolyte used in this study was 0.5 mol dm^{-3} NaCl solution which was prepared by dissolving solid NaCl salt in deionized water. In the case of investigation corrosion inhibition effect of green tea extract, electrolyte was prepared by adding 20 ml of the extract in 180 ml of 0.5 mol dm^{-3} NaCl solution. Electrolyte pH was 5.6 without and with addition of green tea extract.

20 g of green tea leaves was weighed with analytical balance, placed in Erlenmeyer flask and extracted with 400 ml of solvent (40% ethanol 60% of deionized water). The flask is equipped with water condenser and the mixture was heated at 75 °C in an ultrasonic bath for 45 minutes. After that the mixture was twice filtered by vacuum filtration through a double filter paper, to remove the smallest solid particles.

Qualitative analysis of the green tea extract was performed on HPLC System HP Model 1090 with diode array detector (DAD). Licosphere RP C18 column was used with length 250 mm, 4.6 mm diameter and 5 μm particles diameter. Spectra were recorded on wave lengths 268 nm, 280 nm, 374 nm, 310 nm and 350 nm. A two solvent gradient elution were performed. The solvent compositions used were (A) water–H$_3$PO$_4$ and (B) acetonitrile HPLC grade. The solvent

composition changed according to the following gradient: 98/2 at 0 min, 98/2 at 3 min, 70/30 at 25 min, 20/80 at 35 min, 2/98 at 40 min and 2/98 at 50 min. The compounds in tea infusion were identified with reference compounds and literature data on the basis of their HPLC retention times.

Electrochemical experiments were conducted after 1 hour immersion in 0.5 M NaCl solution (pH = 5.6) with or without green tea extract. Linear polarization measurements were performed in the range of ±15 mV vs. E_{corr}. Potentiodinamic polarization measurements were performed with potential scan from the most negative potentials (-250 mV vs. E_{corr}) up to the +250 mV vs. E_{corr} with the scan rate of 0.2 mV s^{-1}. For stainless steel samples the upper potential limit for potentiodynamic polarization was +500 mV due to wide area of passivity.

After potentiodynamic polarization, surface area of electrodes was examined with optical microscope Citoval – Carl Zeiss Jena with the magnification of 100 times.

2. RESULTS AND DISCUSSION

Figure 1 shows the chromatogram of green tea extract which is used for corrosion investigation study.
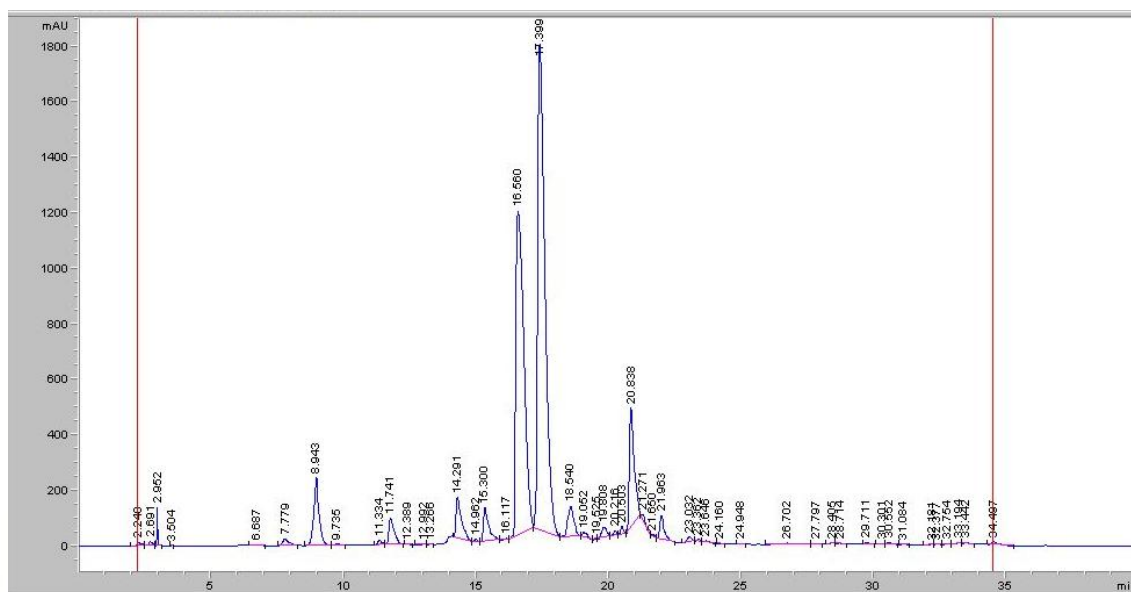

Fig. 1. HPLC chromatogram of phenolic compounds in green tea extract

The main peaks in grean tea extract were from following compounds: epigallocatechin gallate (retention time R_t = 17.399 min), epicatehine (R_t = 16.560 min), myricetin (Rt = 20.838 min) and epigallocatechine (Rt = 8.943 min).

Linear polarization measurements were performed in order to determine the values of polarization resistance of copper, aluminium and AISI 316L stainless steel in 0.5 mol dm^{-3} NaCl solution, without and with addition of green tea extract. Results of these investigations at 25 ^{o}C are shown on Figures 2-4.
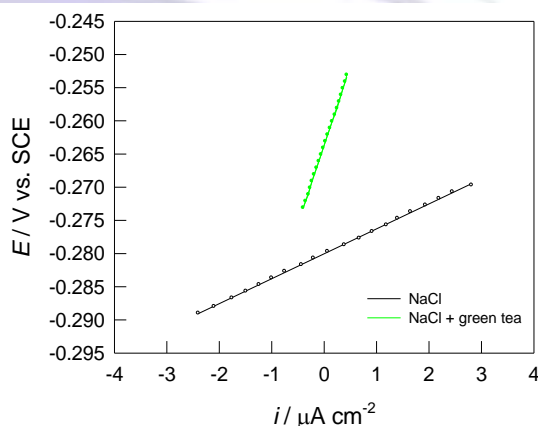

Fig. 2. 'Linear' polarization curves for polarization resistance determination of Cu in 0.5 mol dm^{-3} NaCl solution without and with addition of green tea extract
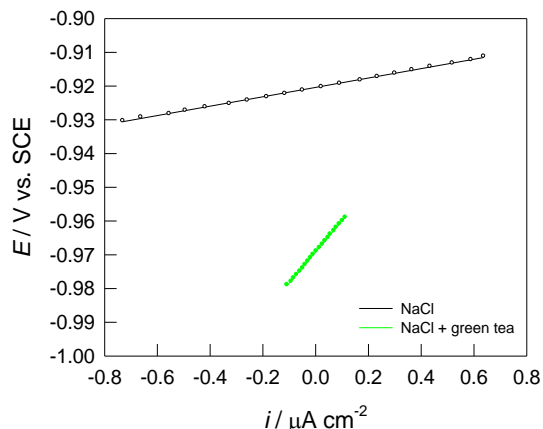
Fig. 3. 'Linear' polarization curves for polarization resistance determination of Al in 0.5 mol dm^{-3} NaCl solution without and with addition of green tea extract
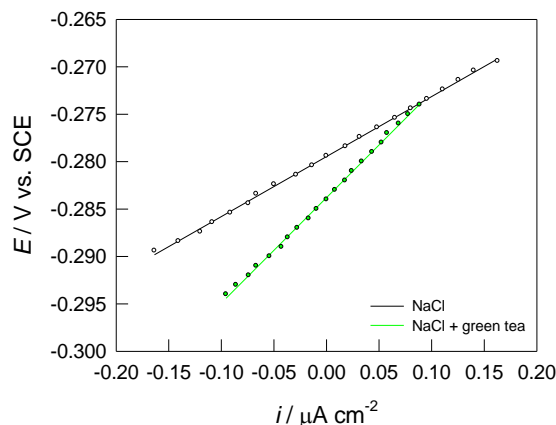
Fig. 4. 'Linear' polarization curves for polarization resistance determination of AISI 316L in 0.5 mol dm^{-3} NaCl solution without and with addition of green tea extract

The values of polarization resistance were determined form the slope of linear parts of polarization curves, in the vicinity of E_{corr}. Straight lines were obtained in the potential range ±10 mV from E_{corr}. R_p values were used to calculate the surface coverage (Θ), and the inhibiting efficiency (η), using the equations:

$$\Theta = \left(\frac{R_{pi} - R_p}{R_{pi}} \right) \tag{1}$$

$$\eta = \Theta \times 100 \tag{2}$$

where R_p and R_{pi} are the polarization resistances without and with the addition of inhibitor.

The values of polarization resistance, surface coverage and the inhibition efficiencies are shown in table 1:

Table 1. The values of polarization resistance, surface coverage and the inhibition efficiencies for Cu, Al and AISI 316L in in 0.5 mol dm^{-3} NaCl solution without and with green tea extract at 20 ^{o}C

metal	solution	R_p / kΩ cm^2	θ	η / %
Cu				
	NaCl	3.758	-	-
	NaCl + 20 ml green tea extract	23.877	0.8426	84.26
Al				
	NaCl	13.916	-	-
	NaCl + 20 ml green tea extract	92.898	0.8502	85.02
AISI 316L				
	NaCl	63.275	-	-
	NaCl + 20 ml green tea extract	111.109	0.4305	43.05

It can be seen that R_p values for Cu, Al and AISI 316L increased with the addition of green tea extract in NaCl solution, which indicates its corrosion inhibition activity. The largest increase in R_p value with addition of green tea extract was obtained for Al and Cu, while this change is much smaller for AISI 316L due its higher natural corrosion resistance compared with Cu and Al.

Potentiodynamic polarization curves were recorded to obtain information about the influence of green tea extract on anodic and cathodic processes on Cu, Al and AISI 316L electrodes in 0.5 mol dm^{-3} NaCl solution.

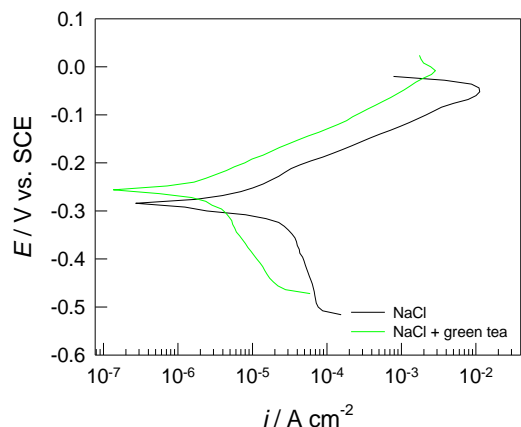The results of potentiodinamic polarization measurements are presented in Figures 5-7:

Fig. 5. Potentiodynamic polarization curves for Cu in 0.5 mol dm^{-3} NaCl solution without and with green tea extract
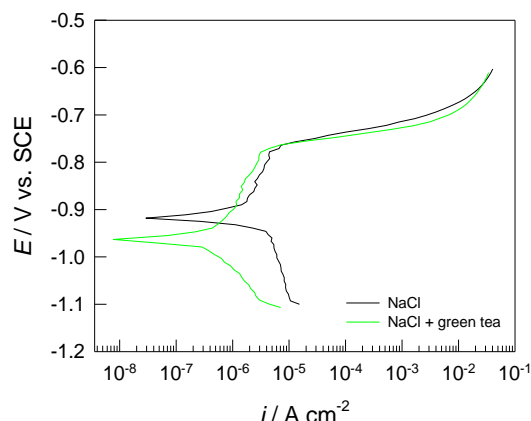

Fig. 6. Potentiodynamic polarization curves for Al in 0.5 mol dm^{-3} NaCl solution without and with green tea extract
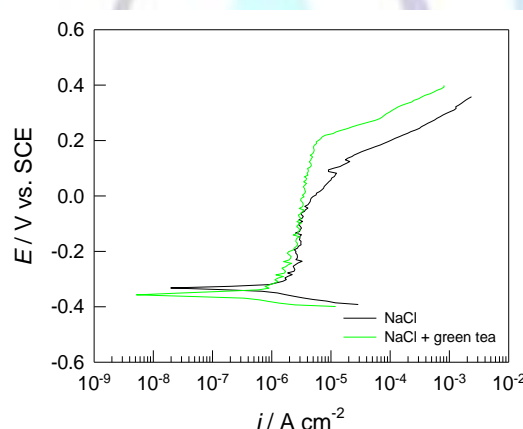

Fig 7. Potentiodynamic polarization curves for AISI 316L in 0.5 mol dm^{-3} NaCl solution without and with green tea extract

Figure 5 shows potentiodinamic polarization curves for copper in 0.5 mol dm^{-3} NaCl solution without and with adition of green tea extract. The cathodic branch of the polarization curve represents reduction of oxygen which can be described by equation (3):

$$O_2 + 2H_2O + 4e^- \rightarrow 4OH^- \tag{3}$$

The anodic branch of the polarization curve represents copper dissolution. The well acepted mechanism of copper corrosion in chloride solutions (when the $c(Cl^-) < 1$ mol dm^{-3}) is two step dissolution process according to equations (4) and (5) [26,27]:

$$Cu + Cl^- \rightarrow CuCl + e^- \tag{4}$$

$$CuCl + Cl- \rightarrow CuCl_2^- \tag{5}$$

In the Tafel-type active region, anodic slope was around 60 mV/decade, which indicates that the polarization behaviour of this alloy was determined by the dissolution of copper to soluble cuprous chloride ion complex ($CuCl_2^-$) and its diffusion to the bulk solution. At more positive potentials (E > -0.05 V), the current density decreases to some extent due to formation of the corrosion products on the electrode surface, which have some protective effect and reduce the active dissolution of metals from the surface. According to the literature, this decrease in current density is usually explained by formation of cuprous chloride (CuCl) and cuprous oxide (Cu$_2$O), surface adsorbed precipitates [27-29]. This layer has very limited protected properties and breaks down with increasing the anodic potential. Thus, the use of corrosion inhibitors in these conditions is justified because on the Cu surface a true protective passive film does not formed.

From the Figure 5. it can be seen that the addition of 20 ml of green tea extract leads to a reduction of anode and cathode current density, and lead to a slight shift of corrosion potential in positive direction in relation to the investigation in a 0.5 mol dm^{-3} NaCl solution without extract. These changes in the polarization curves show that green tea extract acts as a mixed corrosion inhibitor for Cu in NaCl solution.

In contrast to copper, aluminum is a metal that is most easily oxidized which result that the surface of aluminum is almost always covered with the protective oxide film [3]. When the aluminum is immersed in a neutral aqueous solution containing dissolved oxygen, cathodic reaction will be reduction of oxygen, while anodic reaction could be described by the equations (6-8):

$$Al_{(s)} + OH^- \rightarrow Al(OH)_{ads} + e^- \tag{6}$$

$$Al(OH)_{ads} + OH^- \rightarrow Al(OH)_{2ads} + e^- \tag{7}$$

$$Al(OH)_{2ads} + OH^- \rightarrow Al(OH)_{3ads} + e^- \tag{8}$$

The formation of an adsorbed Al(OH)$_3$, which slowly transform to the Al$_2$O$_3$ x 3H$_2$O in neutral solutions result with the passivity of aluminum. Corrosion of aluminum in neutral chloride solutions start with adsorption of chloride ions on sites of irregularities in the surface oxide film [3], which leads to following reactions (9-11):

$$Al_{(s)} + Cl^- \rightarrow AlCl_{ads} + e^- \tag{9}$$

$$AlCl_{ads} + Cl^- \rightarrow AlCl_{2ads} + e^- \tag{10}$$

$$AlCl_2 + Cl^- \rightarrow AlCl_3 + e^- \tag{11}$$

Formation of soluble aluminum chloride compounds lead to dissolution of the oxide film at certain spots and the occurrence of pitting corrosion.

Figure 6 shows potentiodinamic polarization curves for aluminium in 0.5 mol dm^{-3} NaCl solution without and with adition of green tea extract. The anodic part of polarization curve for Al in 0.5 mol dm^{-3} NaCl solution exibited active-pasive-transpasive behavior which is characteristic for existance of oxide film on electrode surface. The pasive area extended from -0.89 V to cca. -0.75 V. Further potential changes in positive direction leads to destruction of the oxide film and to a considerable rise of anodic current. Addition of green tea extract to NaCl solution leads to changes in the polarization behavior of aluminum that are reflected in changes of corrosion potential to a chatodic direction, significant reduction of cathodic current density as well as a small increase in the passive area in the anodic part of the curve. This difference in the corrosion behavior of aluminum in the presence of green tea extract shows that green tea primary acts as a cathodic corrosion inhibitor for aluminum in NaCl solution.

Corrosion resistance of stainless steel is related to the formation of an insoluble, relatively unreactive chromium oxide–hidroxide enriched passive surface film that forms naturally in the presence of oxygen [30,31]. Nevertheless, stainless steels are susceptible to localized form of corrosion in the presence of chloride ions, especially pitting corrosion. Figure 7 shows potentiodinamic polarization curves for AISI 316L in 0.5 mol dm^{-3} NaCl solution without and with adition of green tea extract. The anodic part of the curves shows passive beahvior which is characteristic behavior of stainless steel in neutral aerated solutions [32,33]. At the potential that are more positive than 0 V the breakdown of the passive oxide layer occures. Addition of 20 ml of green tea extract, leads to an increase in the width of the passive areas, as well as a small decrease in cathode current density, which indicates that green tea acts as a mixed corrosion inhibitor for stainless steel. In this case the destruction of the oxide layer evolved at potentials more positive than 0.215 V.

Table 2 shows the electrochemical polarization parameters for the investigated metals in a NaCl solution without and in the presence of green tea extracts. Parameters include corrosion potential (E_{corr}), corrosion current density (i_{corr}), percentage of inhibition efficiency (η), and surface coverage (Θ). Percentage of inhibition efficiency and surface coverage was calculated using equations (12) and (13):

$$\Theta = \frac{i_{corr} - (i_{corr})_i}{i_{corr}} \tag{12}$$

$$\eta = \frac{i_{corr} - (i_{corr})_i}{i_{corr}} \times 100 \tag{13}$$

where i_{corr} and $(i_{corr})_i$ are corrosion current densities in the absence and presence of inhibitor.

Table 2. Corrosion current density, corrosion potential, surface coverage and inhibition efficiency for the Cu, Al and AISI 316L in 0.5 mol dm^{-3} NaCl solution without and with addition of green tea extract

solution	metal	E_{corr} / V	i_{corr} / μA cm^{-2}	θ	η / %
NaCl	Cu	-0.275	24.0	-	-
NaCl + 20 ml green tea		-0.260	2.65	0.8889	88.89
NaCl	Al	-0.920	3.90	-	-
NaCl + 20 ml green tea		-0.961	0.37	0.9051	90.51
NaCl	AISI 316L	-0.334	0.30	-	-
NaCl + 20 ml green tea		-0.360	0.16	0.4666	46.66

From the Table 2 it can be seen that the highest corrosion inhibition efficiency green tea extract showed on aluminum and copper while corrosion inhibition efficiency on AISI 316L steel is relatively small.

After potentiodynamic polarization measurements the electrode surfaces was examined with the optical microscope with a magnification of 100 times. Figure 8. shows the surface of the Cu electrode after potentiodynamic polarization in NaCl solution and Figure 9. the surface of the Cu electrode after potentiodynamic polarization in NaCl solution with addition of green tea extract.

Fig. 8. Optical micrograph of Cu electrode surface after potentiodynamic polarization in 0.5 mol dm^{-3} NaCl solution
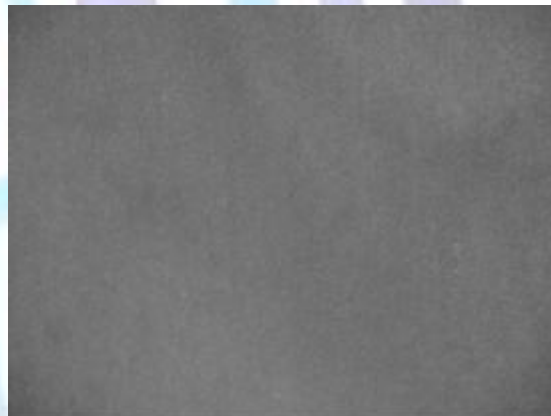

Fig. 9. Optical micrograph of Cu electrode surface after potentiodynamic polarization in 0.5 mol dm^{-3} NaCl solution with adition green tea extract

From the Figures it can be seen that in the case of copper corrosion in NaCl solution, there is no localized corrosion process. The surface roughness of the electrode (Fig. 8) sugest the evolution of general corrosion. Adition fo green tea extract reduces the surface roughness of electrode propably due to adsorption of organic compounds to electrode surface.

On the surface of the Al electrode it can be seen a local corrosion damages (Figure 10), which is propably the result of pitting corrosion, which is the characteristic form of corrosion attack on aluminum and its alloys in a neutral chloride solutions. The presence of green tea leads to a reduction of the corrosion demages (Figure 11).
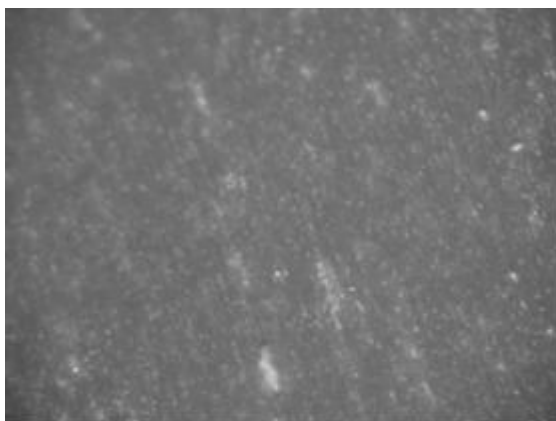
Fig. 10. Optical micrograph of Al electrode surface after potentiodynamic polarization in 0.5 mol dm^{-3} NaCl solution

Fig. 11. Optical micrograph of Al electrode surface after potentiodynamic polarization in 0.5 mol dm^{-3} NaCl solution with adition green tea extract

The surface of the AISI 316L has at least corrosion damage, compared to Cu and Al electrodes (Figure 12 and 13). Small corrosion damages are probably resulting of pitting corrosion. Green tea extract reduces the corrosion damages on steel surface (Figure 13).

Fig. 12. Optical micrograph of AISI 316L electrode surface after potentiodynamic polarization in 0.5 mol dm^{-3} NaCl solution
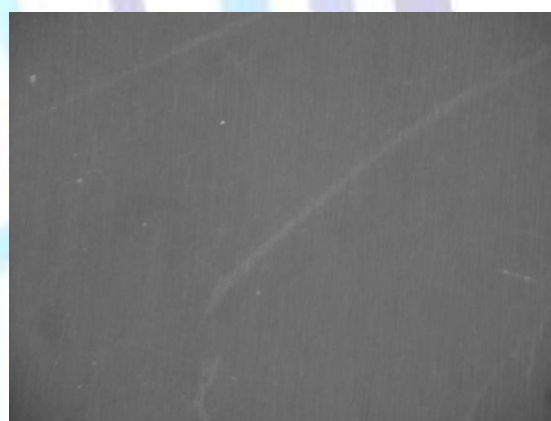

Fig. 13. Optical micrograph of AISI 316L electrode surface after potentiodynamic polarization in 0.5 mol dm^{-3} NaCl solution with adition of green tea extract

3. CONCLUSIONS

- By examining the influence of green tea extract on the corrosion behavior of Cu, Al, Sn and stainless steel AISI 316L in 0.5 mol dm^{-3} NaCl solution, by potentiodynamic polarization method and linear polarization method, it was found that green tea extract acts as a corrosion inhibitor for investigated metals.

- The effectiveness of corrosion inhibition is different for different metals. The highest efficiency of inhibition green tea extract showed on aluminum, followed by copper, while the corrosion inhibition effect of the extract on AISI 316L stainless steel was relatively low ($\eta < 50\%$).

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