



Examining the Corrosion Behavior of 6061-T6 Al Alloy Inside Seawater with Decorative Gold- and Silver-Color Coating

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Highlights

- 6061-T6 Al ALLOY corrosion was examined in sea water
- Gold and silver color organic resin paint was used for coating
- It was determined that the silver color paint had higher corrosion resistance
- Damaged surfaces have lower corrosion resistance than undamaged

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Abstract

This study examined the corrosion resistance of the metallic paint coated, uncoated and damaged paint coated form of the high-strength 6061-T6 Al alloy inside seawater. Solvent-based paint containing two different metallic pigments produced with an alkyd binder was produced and the coating of Al 6061-T6 alloy was made with this paint. To determine the course of corrosion electrochemical impedance spectroscopy method was used. Ecor and Rp values were calculated from potential and current change values. As a result, it was determined from the Ecor, Rp, SEM -EDX images, and Nyquist curves that the corrosion resistance of impact coatings was lower. The corrosion resistance of gold color (Ecor -0.66320 V, Rp 60030.22 ohm) paint substantially containing copper pigment was lower than the corrosion resistance of silver color paint substantially containing Al pigment (Ecor -0.72912 V, Rp 22264.05 ohm) was observed.

1. INTRODUCTION

Metal materials are indispensable elements with a wide range of uses ranging from kitchens to space shuttles to facilitate the life of human beings. The biggest problem that arises during the storage and use of metals is the problem of corrosion. Corrosion, along with natural disasters, can cause great economic losses in essential structures such as watercraft, pipelines, bridges, and buildings [1]. Aluminum alloy 6061 is a typical alloy containing alloying elements Mg-Si-Cu. It is used in both T4 (solution treatment and natural aging) and T6 (Peak aging) temper conditions [2]. One of the places where 6061 Al alloys are widely used in the marine environment. They have advantages such as low density, easy processing, cost-effectiveness, fatigue, and corrosion resistance [3]. Aluminum used in ships is not resistant to corrosion caused by high speed. Among the Al 6061 alloys, T6 alloy is more resistant to seawater and corrosion in this environment [4]. Various methods are used to protect the substrate from the abrasive corrosion of metallic materials. For these, dipping, spraying, chemical or electrodeposited organic-based, metallic and ceramic coating methods are used [5]. Seawater studies are intensified on Al 6061 alloys in the marine environment, chloride ions strongly attack the oxide film by absorbing chemical or physical heterogeneities on the surface of the oxide film. and cause different types of corrosion [6]. Therefore, the most important factor limiting the use of materials in larger areas is corrosion [7]. In order to increase the corrosion resistance of alloys, it is necessary to improve the mechanical and tribological properties [8]. Due to the strong self-passivation, it is more susceptible to crevice corrosion in a neutral solution containing Cl⁻. In view of this, Al alloys are corrosion exposed in the marine environment and crevice corrosion is inevitable [9]. Anti-corrosion coatings are often applied to serve as functional barriers for offshore structures in a variety of corrosive

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environmental environments such as immersion in water or sea, treatment with hot corrosive liquids, burial in soil, and exposure to areas containing industrial heavy chemicals [10]. Coatings for protecting metallic sublayers such as steel, iron, magnesium, and aluminum and their alloys against corrosion have received much attention in the high seas industry [10]. An organic coating provides effective corrosion protection when the coated metal is exposed to a sea environment that is rich in chloride ions [11]. There is usually a need for a long time to determine the corrosion protection provided by coatings that contain organic binders [12]. Especially for calculation and replicability of the numerical value of the corrosion protection of organic coatings, highly significant test methods have been developed [13, 14].

Coatings containing metallic effects have started to be used on metal surfaces not only to prevent corrosion but also for decorative purposes [15, 16]. In addition to this, electrochemical methods have been used for years to measure the coated corrosion resistance of metal surfaces [17]. So far, several studies have been conducted to develop new corrosion inhibitors to protect Al alloys [18, 19]. Many studies were proven that the corrosion resistance of aluminum alloys is higher than that of pure aluminum [20].

In this study, the corrosion behavior of paints containing metallic pigments, which can be used for decorative purposes for marine vessels, in seawater was investigated. Two different fast-drying alkyd-based paints containing metallic pigments were prepared in the laboratory environment. These paints included pigments that provided the colors of gold (substantially Cu pigment) and silver (substantially Al pigment). The decorative usability of such paints in environments that usually have contact with seawater and their contribution to the corrosion resistance of the 6061-T6 Al alloy inside seawater were examined. The corrosion behavior of the damaged surfaces of alkyd resin-based paint coatings incorporating metallic pigments weren't investigated in scientific investigations. This study will provide an understanding of how damaged painted surfaces are affected by seawater corrosion. On the other hand, the corrosion behavior of paints produced from two different metallic pigments was not investigated at the same time in the literature. This article was written to draw attention to using paints containing metallic-colored pigments to improve corrosion damage prevention methods.

2. MATERIAL METHOD

2.1. Material

In this study, as the experiment specimens, 6061-T6 Al alloy specimens with diameters of 3 mm and lengths of 70 mm were utilized. The chemical composition of the 6061-T6 Al obtained from the firm Seykoç is given in Table 1, while its mechanical properties are shown in Table 2.

Table 1. 6061-T6 Al Chemical Composition

Al	Fe	Si	Cr	Mn	Mg	Zn	Cu	Ti	Other
Remainder	0.7	0.4-0.8	0.04-0.35	0.15	0.8-1.2	0.25	0.15-0.40	0.15	0.15

Table 2. 6061-T6 Al Mechanical Properties

Temper	Yield Strength (MPa) min-max	Tensile Strength (MPa) min-max	Elongation (50%) min-max	Hardness (Brinell) min-max
T6	240-270	260-310	20	95

The KCl, HNO₃, H₂SO₄, technical-grade acetone, and ethanol that were used for material cleaning were obtained from the firm MERCK. In coating, the parts of all included electrodes that were not used for measurements, a polyester resin obtained from the firm TİLA KOMPOZİT was used. In preparation for the internal solution of the reference electrode (3M KCl), ultra-distilled water was used as the solvent. Genuine seawater collected from Burhaniye in the Balıkesir province of Türkiye was used in the experiments. The paints that were used as a coating to test the corrosion resistance of 6061-T6 Al were produced in laboratory conditions, and their contents are given in Table 3. The materials that were used in

paint production was obtained from the firm ASİL KİMYA VE BOYA SAN. VE TİC. A.Ş. The metallic pigments that were used were in a layer-forming structure.

Table 3. Contents of paints used in corrosive medium

	Raw Material Name	Gold color %	Silver color %
1	Styrenated Alkyd 65%	70	70
2	Texaphor 963	0.2	0.2
3	Bentogel BG4	0.2	0.2
4	Toluene	25.5	25
5	Au Color lifing Paste	3.5	-
6	Ag Color lifing Paste	-	4
7	Dryer Calcium Octate 4 %	0.4	0.4
8	Anti Incrustation MEKO (C ₄ H ₉ NO)	0.2	0.2

A Silver/Silver Chloride electrode (Ag/AgCl, 3 M KCl) was used as the reference electrode. A Platinum plate (10 mm x 10 mm x 0.1 mm) was used as the counter electrode. The total electrode surface area was 2 cm². All surfaces for corrosion examinations were coated with polyester so that only the parts that would contact the test solution would stay uncoated. Nitrogen gas was used to try the surfaces of the Al electrodes. A Mechanical Polisher (Imro Propol-VTD) was used to polish the surfaces of the electrodes. After the Al electrodes were polished, an Ultrasonic Bath (Elma S60H Elmasonic) was used to remove and clean the polishing residues and metal particles on the surface. A Potentiostat-Galvanostat (CHI 6096E) was used in the electrochemical measurements. A Scanning Electron Microscope (SEM) (JEOL 6510) was used to examine the electrode surfaces. Energy-Dispersive X-Ray Spectroscopy (EDX) (JEOL 6510) was used in the elemental analyses of the surfaces of the Al electrodes that were treated with the corrosive medium. A Metal Saw was used to cut the Al electrodes. Sandpapers in various grades were used to polish the surfaces of the 6061-T6 Al electrodes. The experiments were carried out at a constant temperature of 25°C.

2.2. Method

The fast-drying gold- and silver-colored paints that were used as corrosion inhibitors were produced in the laboratory environment with the contents shown in Table 3. Their drying times were determined as 35±2 minutes. The viscosity of the produced paints was determined as 75±5 sec. The corrosion experiments were conducted inside seawater. Before usage, the seawater was filtered through a simple filter paper.



Figure 1. Polymer coating of the side surfaces of the specimens

All 6061-T6 Al alloy specimens were prepared in the form of rods with a diameter of 7 mm and a length of 5 cm. To provide a smooth surface on the prepared rods, the parts that would be subjected to corrosion were thoroughly sanded. One end was drilled and equipped with a copper wire to achieve electrical conductivity as shown in Figure 1, the end contacting the test solution was not drilled, and in a way to allow the other end to stay in the open, all other parts were coated with polyester.

The paint was sprayed onto the surface to be subjected to corrosion from a distance of 25 cm. The specimen was left for 1 day so that there would be no problem regarding the drying process. The paint thickness was about 250 ± 5 μm for all specimens. The fast-drying gold- and silver-colored paints that were used as corrosion inhibitors were produced in the laboratory environment with the contents shown in Table 3. Their drying times were determined as 35 ± 2 minutes. The viscosity of the produced paints was determined as 75 ± 5 sec. The corrosion experiments were conducted inside seawater. Before usage, the seawater was filtered through a simple filter paper.

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2.3. Electrochemical Measurements

All corrosion experiments were conducted electrochemically with the 3-electrode technique. The prepared surfaces were used as the working electrodes, the Pt counter electrode, and the Ag/AgCl, 3M KCl reference electrode. The measurements were carried out with a computer-controlled CHI 660D Galvanostat device. The measurements were made in an open atmosphere, and the solution temperature was kept constant at approximately 25°C.

The working electrodes were submerged into the working test solutions, and the change in the open circuit potential for 1400 seconds (E_{ocp-t}) was graphically recorded. From the obtained changes, it was aimed to acquire information on the start and progress of the corrosion, as well as which of the anodic or cathodic reactions affected the corrosion and how. After the open circuit potential reached an equilibrium in 1400 seconds, electrochemical impedance spectroscopy (EIS) measurements were made in the frequency range of 100 kHz and 0.003 Hz by applying an amplitude of 5 mV.

The linear polarization resistance (LPR) determination measurements were made with the potentiodynamic method at ± 10 mV from the open circuit potential. The scanning rate in these measurements was 1 mV/s. The polarization resistances were calculated from the slope of the current-potential curves that were graphically obtained. Regarding the open circuit potential, the analysis of the anodic and cathodic potentials was carried out in the range of +1.8, -1.8.

2.4. Characterization of Electrode Surfaces

After keeping the Al electrodes in seawater, the structure of their surfaces was examined by SEM. The surface composition of the metal was determined by an EDX device combined with the SEM device. By placing the electrodes horizontally in the SEM device, the layer thicknesses of the painted electrodes were determined.

3. RESULTS AND DISCUSSION

To examine the effect of the coating on the corrosion surface, in the study, uncoated corrosion of 6061 T6 Al aluminum alloy was first investigated. Following this, the corrosion behavior of silver-colored and gold-colored paints and their impacted states were investigated and compared. Experiments were carried out for different surface conditions of 6061-T6 Al alloy material at 2000 mm/s scanning speed, until the moment of reaching equilibrium in seawater at room temperature for 1400 seconds. Based on the data obtained from

Figure 2, the corrosion potential E_{cor} and the total resistance polarization resistance (R_p) values on the metal surface were calculated with the Tafel Extrapolation Method and presented in Table 4.

Table 4. E_{cor} values of the uncoated, coated and coated/damaged forms of the 6061 T6 Al alloy

Material surface	E_{cor} (V)	R_p (ohm)
6061-T6 Al Alloy	-0.72912	22264.05
6061-T6 Al Alloy– silver-color-coated	-1.0479	1194384.9
6061-T6 Al Alloy– silver-color-coated damaged	-0.93892	599766.2
6061-T6 Al Alloy– gold-color-coated	-0.66320	60030.22
6061-T6 Al Alloy– gold-color-coated damaged	-0.66244	16749.10

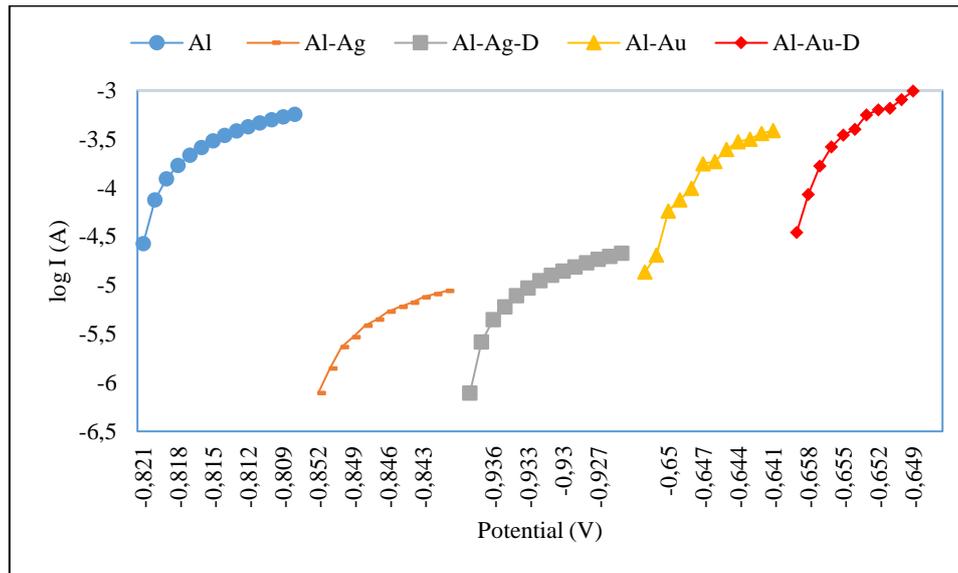


Figure 2. Cathodic side potential current graphs of all specimens

The open circuit potential is the potential value measured while there is no current passing through the circuit. It is one of the common methods used to monitor corrosion development [21]. To examine the effect of the coating on the corrosion surface, the corrosion current obtained from the Tafel curves is a decisive parameter [22]. According to the Faraday equation, the current density is one of the parameters used to determine the corrosion rate. In this relation, there is a direct proportionality between the amount of current passing through the material while the corrosion reaction continues and the amount of solute from the material. According to the Faraday equation, the low current density during the corrosion of the material indicates the high corrosion resistance of the material [23]. Table 4 shows that the E_{cor} value of the paint-coated AA6061-T6 Al gives lower values than the E_{cor} value of the bare AA6061-T6 Al in all cases. It is observed that the E_{cor} values of the impacted paints are lower than the non-impact paints. From Table 4, it can be said that the most suitable coating type to prevent corrosion is the alloy coated with silver paint, but Tafel diagrams are not sufficient for such precise statements.

3.1. Electrochemical Impedance Spectroscopy Results

It is known that the EIS method is preferred because it does not disturb the metal/solution interface and the results obtained are reliable [24]. Electrochemical impedance spectroscopy technique was applied in this study. One of the easiest ways to study the corrosion of coatings and bare metals in seawater is the Electrochemical impedance spectroscopy EIS technique. This technique was widely used recently in the investigation of metal surfaces, especially in corrosion studies. In this technique, a small amplitude alternating current (or potential) is applied to the metal and the behavior of the metal is studied. Since a linear current (or potential) is not applied, the surface is not polarized and distorted. This provides enormous advantages. Since the small amplitude alternating current applied to the metal surface does not change the surface structure much, more accurate results can be obtained regarding the resistance and surface structure

of the metal. The resistance determined by this method is the polarization resistance, in addition to this, the charge transfer resistance, the coating or film resistance on the surface, and the resistances against ion diffusion can affect the system depending on the situation [25, 26].

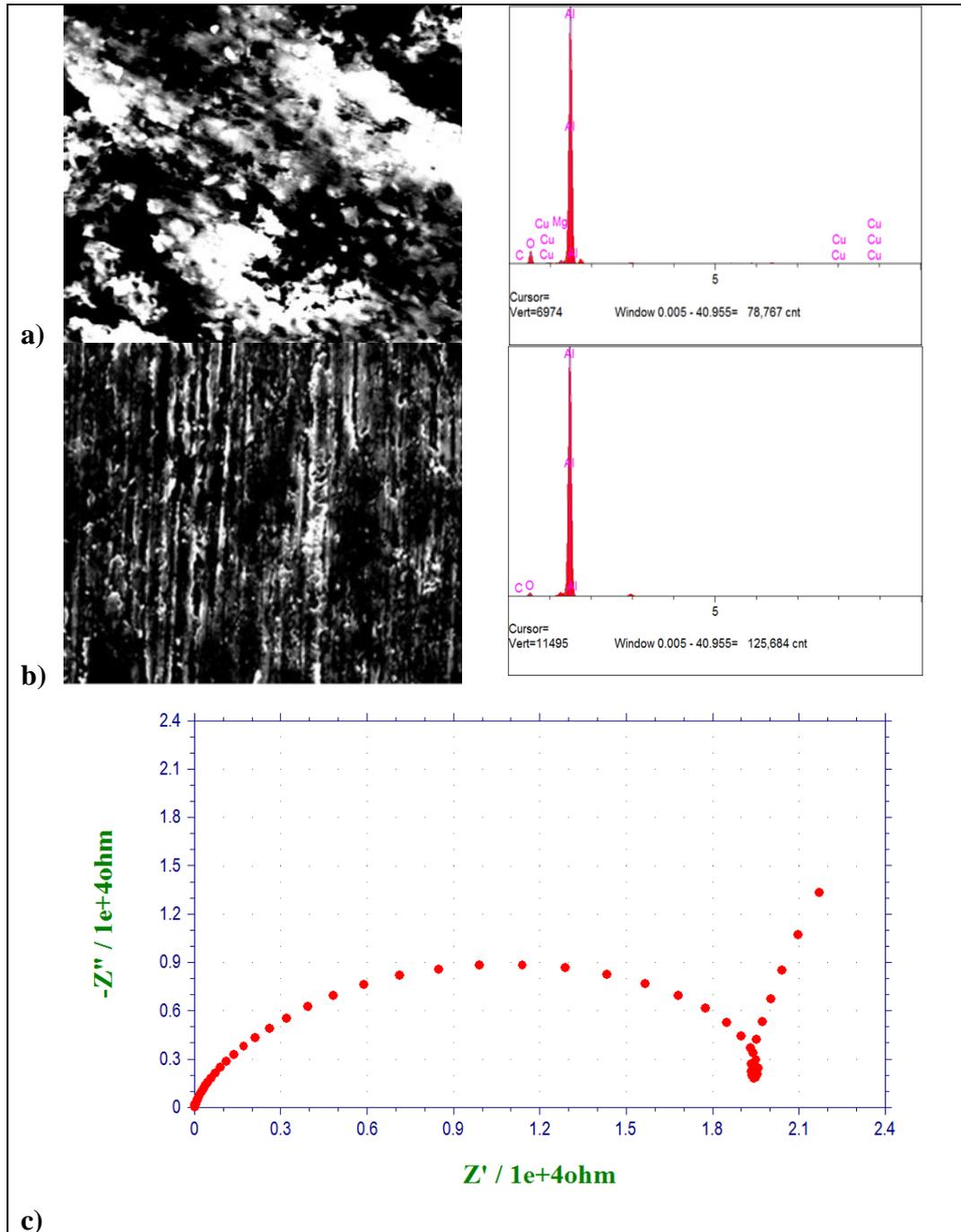


Figure 3. 6061-T6 Al a) SEM -EDX results before corrosion, b) SEM -EDX results after corrosion (kV 20.0, Takeoff Angle 35.0, Elapsed Livetime 10.0), c) 1400-sec Nyquist curve

EIS is widely used in the investigation of electrochemical events occurring at the solution-metal interface. In this method, the polarization resistance can be calculated as the sum of all resistances formed at the metal-solution interface. It also provides important information about the corrosion resistance of corrosive ions on the surfaces of paint or other coatings applied to the surface [27, 28].

Figure 3 presented the corrosion behavior of 6061-T6 Al alloy without paint application. In the a) part of the figure, the corrosion-free carpet and EDX graph of the metal are seen. The (a) and (b) parts of the figure

were examined together, the surface appearance after corrosion shows that there were new chemical deposits on the metal surface. After EDX analysis, it is seen that the Al compounds are in the majority on the surface after the corrosion, and besides, the other ions in the alloy dissolve and accumulate on the surface.

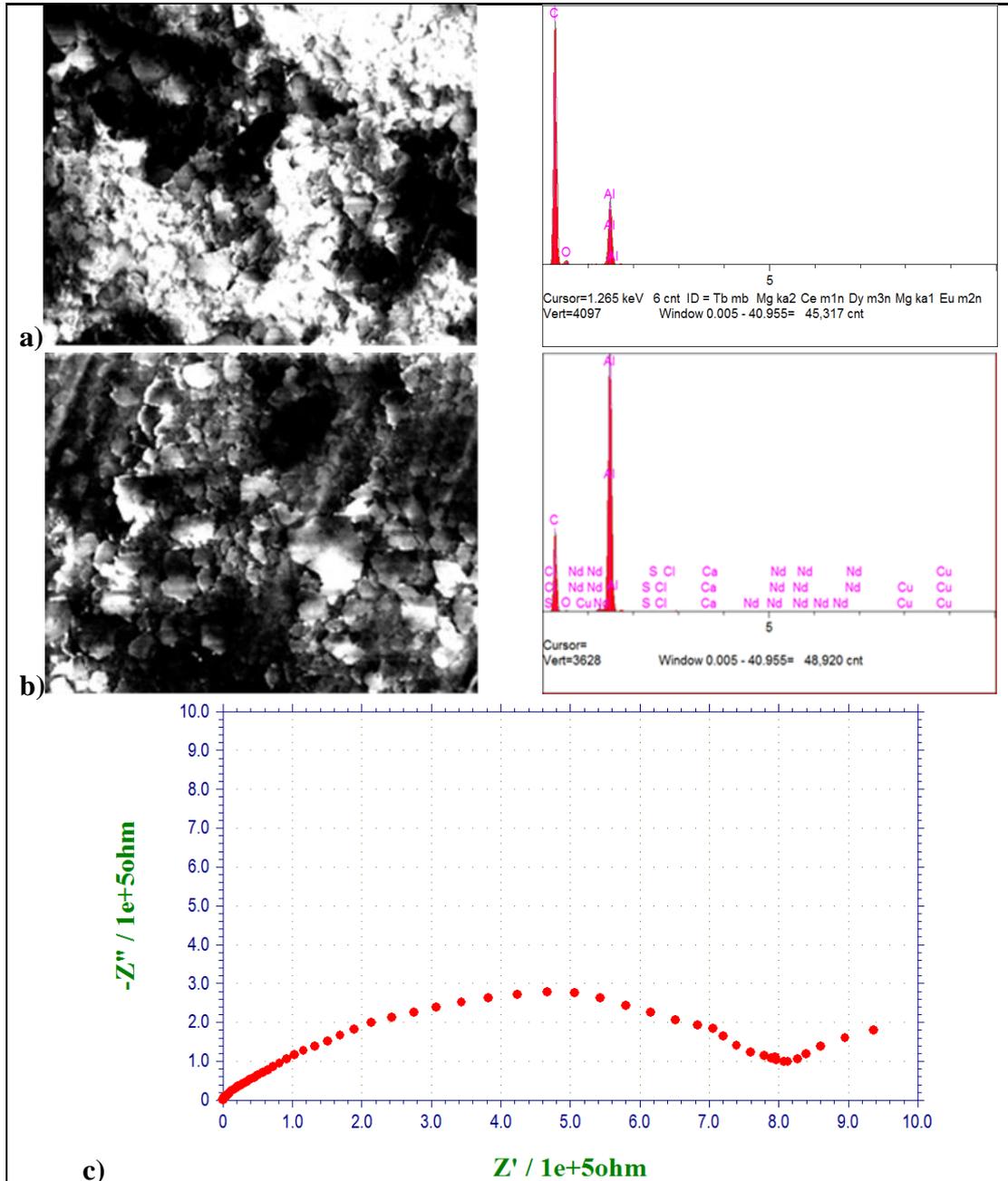


Figure 4. 6061-T6 Al silver-color-coated. a) SEM -EDX results before corrosion, b) SEM -EDX results after corrosion (kV 20.0, Takeoff Angle 35.0, Elapsed Livetime 10.0), c) 1400-sec Nyquist curve

Solvent-based alkyd coatings are known to protect metals against corrosion [29, 30]. High impedance and improved anti-corrosion performance of alkyd copolymer films were confirmed by exposure tests in previous studies [31]. Figure 4 shows the corrosion behavior of paint coated with silver paint. Compared to Figure 3, the difference between the corroded and non-corrosive SEM images and the EDX images were showed that the metal surface is less corroded due to the coating. When the Nyquist curve is examined, the diameter of the curves increases as the corrosion rate decreases due to the coating of the metal surface with paint. This change was showed that the paint covers the metal surface, forms a protective inhibitor film, and reduces the corrosion rate by reducing the number of active centers on the metal surface that come into

contact with the corrosive solution [32]. In Nyquist curves, capacitive loops are associated with charge transfer in the corrosion reaction and the protective film layer formed on the surface. The paint used in this study acted as a protective film layer and reduced the corrosion rate. The area under the capacitive curve obtained from the EIS measurements is an indicator of corrosion resistance. Large area Nyquist curves indicate high corrosion resistance [33]. The SEM image was showed the presence of passive films on the alloy surface and was showed that the corrosion resistance is high without any holes or pits [34].

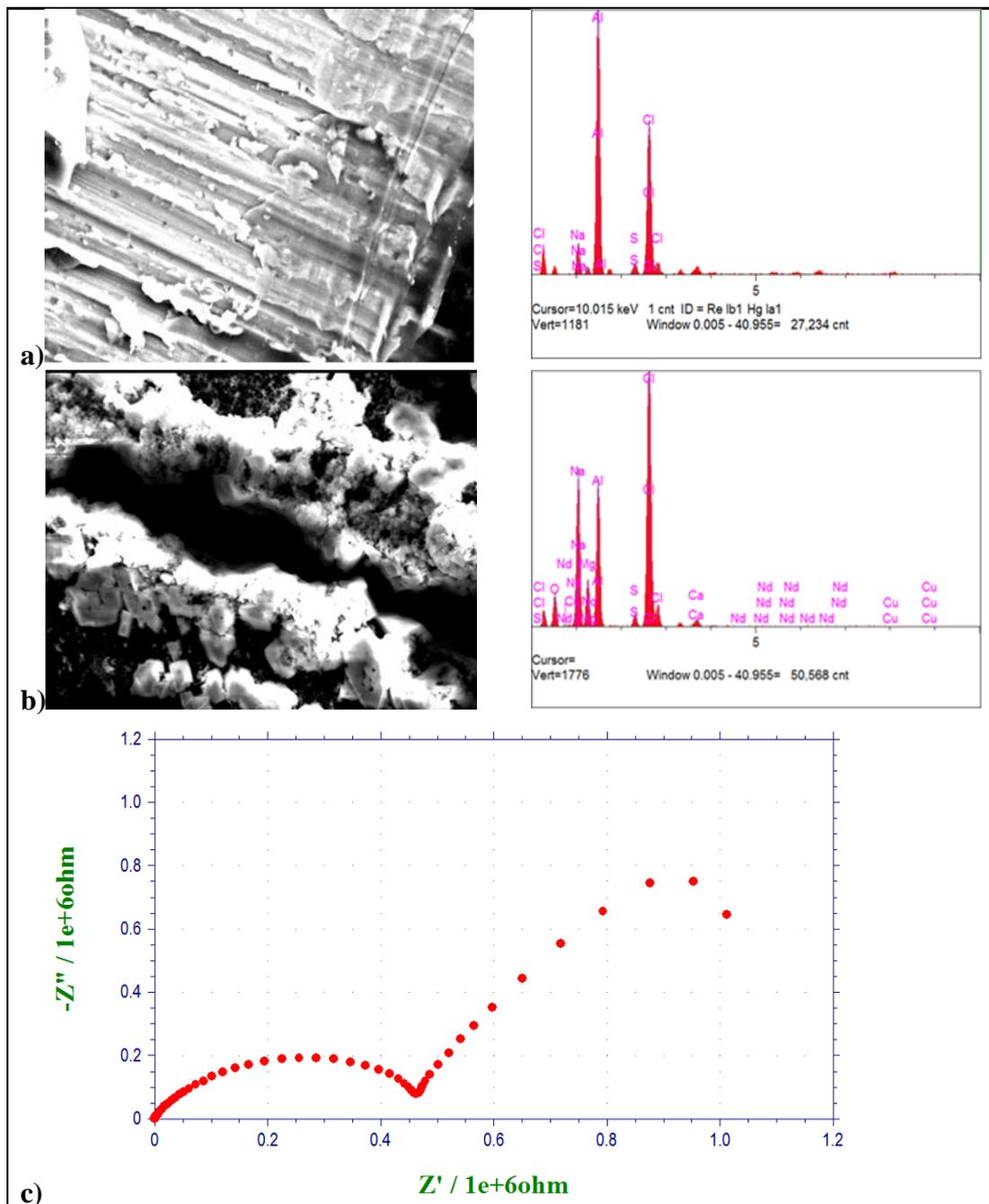


Figure 5. Damaged 6061-T6 Al and silver-color-coated. a) SEM -EDX results before corrosion b) SEM -EDX results after corrosion (kV 20.0, Takeoff Angle 35.0, Elapsed Livetime 10.0) c)1400-sec Nyquist curve

Passive films, conversion coatings, and metallic and organic coatings provide corrosion protection through a variety of mechanisms, including the creation of barriers to penetration of abrasives [35]. When Figure 5 is compared to Figure 4, it is seen that the separation of the paint with damage on the coated surface increases the corrosion. With the EIS method, it is possible to examine the response of an electrochemical

system with small signal effects, and it is a very effective technique where it is possible to identify and analyze any point of the system. [36]. The electrochemical impedance spectra EIS provides detailed information on corrosion reaction mechanisms. Electrochemical impedance spectroscopy (EIS) is a valuable characterization tool for a wide variety of materials due to its ability to probe transport and reaction pathways over a wide range of time scales [25, 26]. As a matter of fact, this study, it was enable us to easily prove how small damage on painted surface increases the rate of corrosion in a short time.

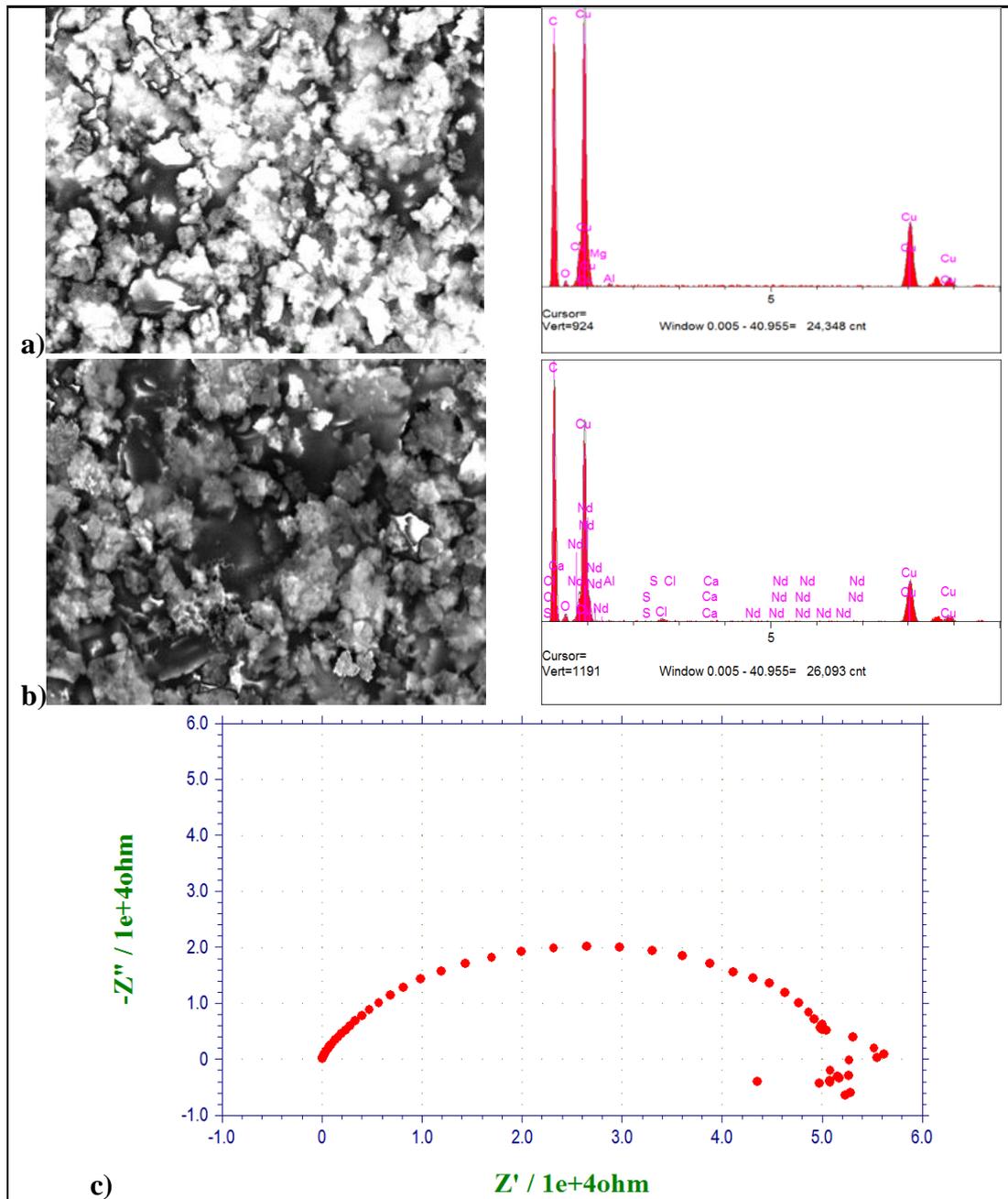


Figure 6. 6061-T6 Al gold-color-coated, a) SEM -EDX results before corrosion, b) SEM -EDX results after corrosion (kV 20.0, Takeoff Angle 35.0, Elapsed Livetime 10.0), c)1400-sec Nyquist curve

In contrast, in sterile seawater, chloride ions were adsorbed at the phase interface boundary, where some of the dissolved chemicals were observed to accumulate. Once the passive film was removed, the metal was directly exposed to the solution, resulting in localized pitting corrosion [37]. The chlorinated compounds formed around the damaged part can be seen very clearly from the figure and EDX. In the disintegrating paint film, some of the water-insoluble ions that can react in seawater were collected at the damaged edge. In the meantime, the presence of ions such as Mg and S in the content of 6061 AL alloy, which is released

as a result of the damage between the EDX made in its non-corrosive form and the EDX with corrosion can be easily observed. It was observed that the binder and aluminum pigments in the paint significantly improved the corrosion resistance [38]. Compared to the coated form, Alkyd coatings prevented corrosion on the bare metal surface before and resulted in the same in this study [30]. It has been observed that the occurrence of corrosion shortens the life of the metal alloy [9]. In the Nyquist curve, the inductive response in the Low-frequency region is related to the adsorption of chloride in the pitted regions during the pitting process [26]. Paints prevent contact at the metal-solution interface. For example, it is known that damaged coatings generally do not provide a normal Nyquist semicircle and their impedance values are lower than undamaged ones because their corrosion resistance is smaller [39].

Figure 6 shows the SEM-EDX diagrams of the surface coated with gold paint with and without corrosion and the Nyquist curve of the corrosion. SEM and EDX images show that the effect of the paint on corrosion was very good and the coating was durable enough not to expose the metal surface during corrosion. When EDX was examined after corrosion, it was seen that copper, which is the main element that creates the gold color, was seen in the foreground and there were C atoms originating from the resin. There were no Al residues on the surface as the main material was not dissolved. It was observed that the Nyquist curves do not form a smooth structure and there was a shift to the negative part of the graph in the Low-frequency Region. The EIS spectrum can be divided into 3 regions. Ohmic region represents the internal resistance of the cell measured where the cell impedance changes from inductive to capacitive behavior [26, 40].

Mid-frequency region: Represents charge transfer processes, both from the electrolyte to the electrode surface and from the electrode surface to the bulk active material of the electrodes. It is always in the form of flat semicircular arches. It usually consists of a single semi-arch or two semi-arches, depending on cell chemistries and/or the specific mechanism of aging going on [41]. Low-frequency region: represents the diffusion processes at the two electrodes [40]. This region is often neglected or even unmeasured, so it can be monitored for the longer time required to measure impedance at lower frequencies [42]. The high-frequency inductive behavior of the cell was neglected in this study as it has little effect on the cell behavior. The semicircle below the (-) axis in the Nyquist plot is represented in the circuit model by an illustrative example element with negative resistance and negative capacitance. [25]. Additional resistances occur as a result of corrosion if there is a transferable build-up in the solution. In such cases, the total resistance is used. If the mineral coating used is not a single type, but a composite material containing various materials such as paint, the resistance increases. The electrochemical impedance spectra EIS provides detailed information on corrosion reaction mechanisms. The EIS method can characterize electrochemical events with Nyquist representation as a function of the real part (Z_r) of the negative ($-Z_j$) of the imaginary part of the impedance obtained at different frequencies. Electrochemical impedance spectroscopy (EIS) is a valuable characterization tool for a wide variety of materials due to its ability to probe transport and reaction pathways over a wide range of time scales [25, 26].

Normally for comparison of damaged and undamaged surfaces in ideal systems, the Nyquist curve should have the appearance of a perfect semicircle. Through the open part of the coating, the solution contacts the metal directly. The charge to the surface between the metal and the solution is transferred by electrons in the metal part, while it is transferred by ions in the solution part. As the diameters of the ions get larger, the ions take up too much space in the solution for the charge that electrons of the same size can provide, and they begin to move deeper into the solution [43, 44].

Figure 7 shows the damaged surfaces of gold color coated SEM, EDX images and Nyquist curve. Residues from dissolution after damaged, such as the gold-colored (containing Cu molecules) damage surface, were accumulated around the damage. It is clearly seen that the optical intensifier Nd compounds emerge as a result of corrosion. Al ions, which were not observed in Figure 6, can be easily seen here. This fact shows the corrosion resistance of the coating. The narrowing Nyquist curve shows that impact increases corrosion. More movement was observed in the minus part of the chart. The phenomenon occurring in this Low-frequency region shows that the dissolved metal ions were not homogeneously distributed in the solution, but collect on the surface and form an unstable structure there.

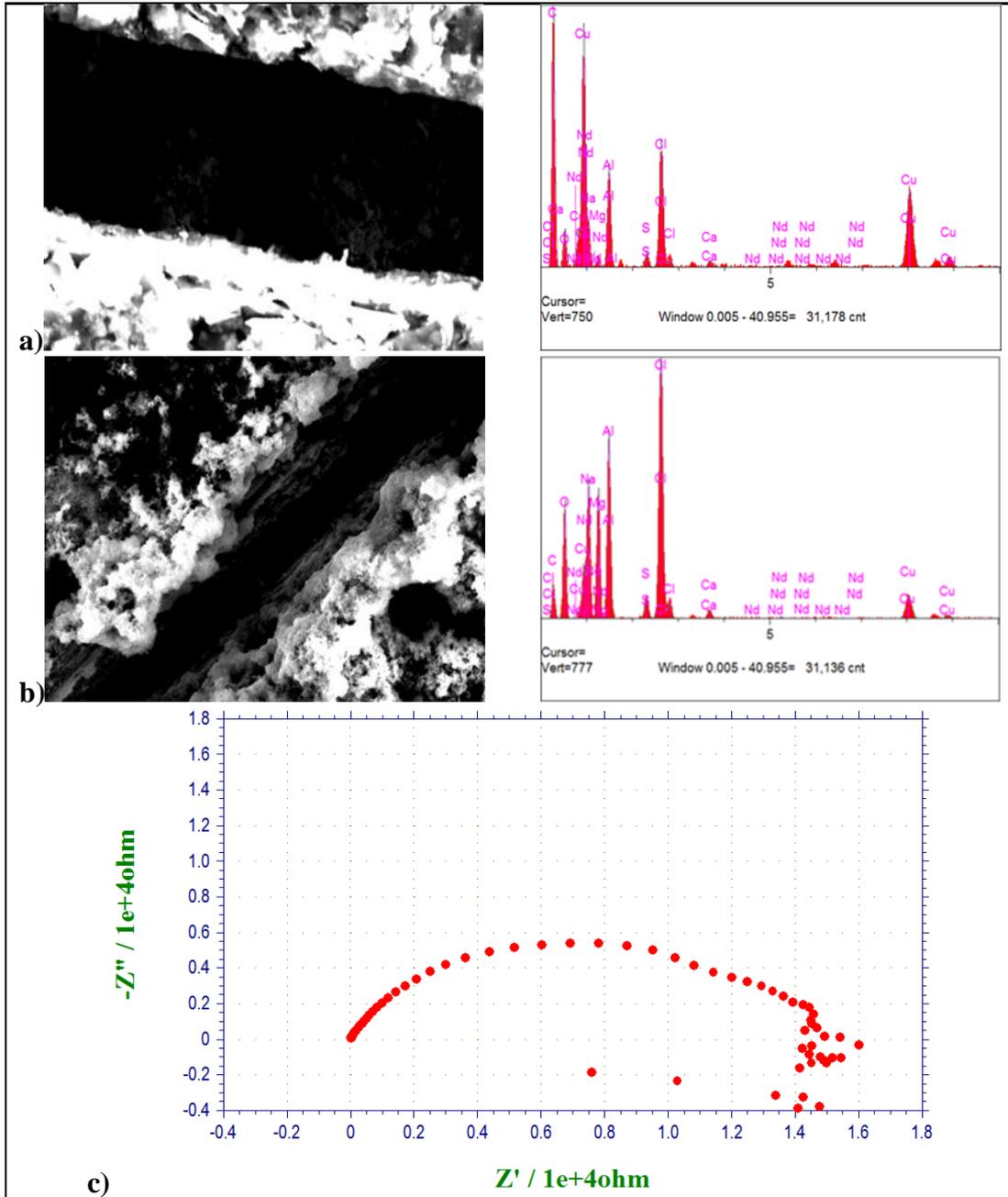


Figure 7. Damaged and 6061-T6 Al and gold-color-coated 6061-T6 Al gold-color-coated, a) SEM -EDX results before corrosion, b) SEM -EDX results after corrosion (kV 20.0, Takeoff Angle 35.0, Elapsed Livetime 10.0), c) 1400-sec Nyquist curve

Pigments in the form of small metallic plates of Al and Cu in paints containing metallic pigments do not show homogeneous distribution. Pigment sizes are not fixed. This inhomogeneous distribution is the reason why the Nyquist curve of coated aluminum was not very regular and the curves show deviations. In addition, as determined from the SEM images, there were problems in the application of gold-colored Cu-containing coated surfaces due to the larger metallic pigment size and in obtaining a smooth surface. This was an expected result. The fact that the pigments of the silver-colored paint were smaller than the gold-colored paint both facilitated the application and enabled us to obtain a smoother coating surface. Therefore, Nyquist curves were exhibited a smoother structure.

4. CONCLUSIONS

In this study, paint was produced using metallic pigments and an organic binder. Corrosion of uncoated 6061-T6 Al alloy and its paint-coated forms in seawater was investigated. In addition, small damages were made on the painted surfaces and the corrosion mechanisms of these damaged surfaces in seawater were investigated. As a result, it was observed that the corrosion resistance of the coated surfaces was higher than the uncoated surface, and the corrosion resistance of the damaged surfaces was lower than that of the undamaged surfaces. It was determined that the number of resistors on the surface increases with the increase in the complexity of the coating content. When Nyquist curve an Rp values were examined, it was seen that the irregularity of Nyquist curves increased and their diameters decreased with increasing resistance. In addition, it was observed that the gold-colored coating containing Cu and Al pigments was less durable than the silver-colored coating containing only Al pigments. It was observed that the resistance was lower (Ecor -0.66320 V, Rp 60030.22 ohm), especially on gold color coated surfaces.

CONFLICTS OF INTEREST

No conflict of interest was declared by the authors.

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