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EFFECT OF TIME VARIATION AND ADDITION OF TARTARIC ACID IN SULFURIC ACID ELECTROLYTE ON PORE THICKNESS AND SIZE OF AL_2O_3 LAYER RESULT OF ROOM TEMPERATURE HARD ANODIZING PROCESS IN THE 1XXX SERIES ALUMINUM MATERIAL

Yoga Nugraha, Rini Riastuti , Pradoto Ambardi, Djoko Hadi Prajitno
Faculty of Engineering, University of Indonesia, Indonesia
General Achmad Yani University (UNJANI), Indonesia
National Research and Innovation Agency (BRIN), Indonesia
E-mail: yoganugraha170816@gmail.com, jasatuti@metal.ui.ac.id, pradambardi@gmail.com, dhppr@yahoo.co.id

Abstract

Hard anodizing is a process that increases the thickness of the Al_2O_3 oxide layer on the aluminum surface, thereby increasing hardness. Factors such as applied voltage, current density, anodized surface area, and anodizing time affect the thickness of the oxide layer. In the research described, aluminum AA 1100 was used and the solution used was sulfuric acid 45 g/L with the addition of $C_4H_6O_6$ (Tartric Acid) 70, 75, and 80 g/L. The hard anodizing process was carried out for 60, 120, and 180 minutes at room temperature (±25°C). The increasing processing time and tartaric acid concentration resulted in a higher thickness, reaching a thickness of 21.37 µm. The XRD results show that the layer compound formed is Al_2O_3 and SEM observations show a porous diameter of 203-408 nm.

Keywords: Hard anodizing, AA 1100, $C_4H_6O_6$ (tartaric acid), XRD, SEM, porous diameter 203-408 nm.

Introduction

The material aluminum is very abundant on earth and at the end of the 19th century became an economic competitor in engineering applications (Bakti et al., 2022) . In 1886, the technology of electrolytic reduction of alumina (Al_2O_3) was discovered by Charles Hall in Ohio and Paul Heroult in France, and this resulted in the appearance of the first vehicle using an internal combustion engine. Aluminum plays an important role as an automotive material that increases engineering value (Majanasastra, 2016) . Within a few decades, the Wright brothers created a thriving industry processing the basic material aluminum. The results of this industrial development are products such as vehicle parts, aircraft bodies, engines, missiles, fuel cells, satellite components and others (Davis, 1990) .

Aluminum material provides a number of advantages when used for production processes, such as an easy manufacturing process and affordable production costs. The mass production process that speeds up production can also be achieved more quickly

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compared to other metals such as titanium, zirconium and other high strength metals. (Manullang & Hutabarat, 2013) . The use of aluminum metal in biomedical products such as implants is more efficient compared to other metals, because aluminum is lighter. The average weight of aluminum is 2.70 g/cm³, while titanium is 4.51 g/cm³ and zirconium 6.52 g/cm³. These advantages make aluminum a material that is highly needed and sought after by the medical application industry (Kurnia et al., 2023) .

Therefore, this research uses aluminum as a base metal material to help applications in the medical or industrial fields that require thin shapes and the formation of micro pores . The aluminum series chosen is the 1xxx series with a content of ≥99.00%. However, due to the thin form of the 1xxx series, aluminum has potential toxicity related to the nervous system, brain, bone disease and risk of anemia, so it is important to reduce its use. Therefore, the hard anodizing process is carried out to thicken the $Al_2O_{3\ \text{layer}}$ on pure metal, which will strengthen and make the product safe for the health sector. (Yusdiana, 2022) .

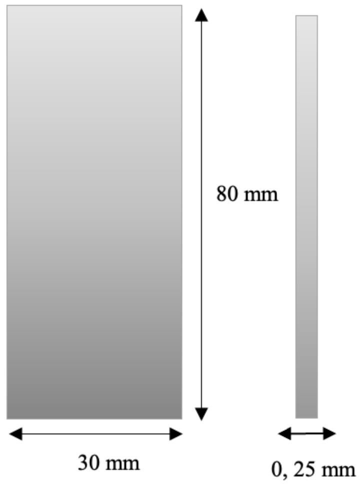
The thickness of the oxide layer in the hard anodizing process can be influenced by several factors such as the voltage applied, the density of the current flowing, the surface area being anodized, the process temperature, and the duration of the anodizing process itself. The quality of the results from the hard anodizing process is usually reflected in the thickness and hardness of the resulting oxide layer (Wood, 1982) . By using a mixture of H2SO4 (sulfuric acid) and $C_4H_6O_6$ (tartaric acid) with a certain concentration and a certain electrical voltage, it is hoped that this process will optimally improve the physical and mechanical properties of aluminum (Mubarok et al., 2016) . To achieve the desired results, research is needed on the anodizing process so that the results of this research can become a reference for the Indonesian industrial world in making products with better quality. Recent advances in the development of biocomposite materials show that alumina (Al_2O_3) can be used for biomedical applications because it is thin and chemically stable, while its biocompatibility can be improved by forming a microporous structure. (Ward et al., 2003) . With this development, apart from being used to harden the $Al_2O_{3\ \text{layer}}$, the hard anodizing process can also be used to create a microporous structure in Al material. This can be achieved by planning the solution, current density, temperature, and duration of the hard anodizing process.

The aim of this research is to determine the effect of electrolyte solution concentration in the hard anodizing process on the thickness of the oxide layer and the formation of micropores. It is hoped to achieve optimal results from the hard anodizing process on 1xxx aluminum alloy and also become a reference for industry, especially small and medium enterprises in this country, to produce high quality products that can compete in the market .

Research methods

1. Material

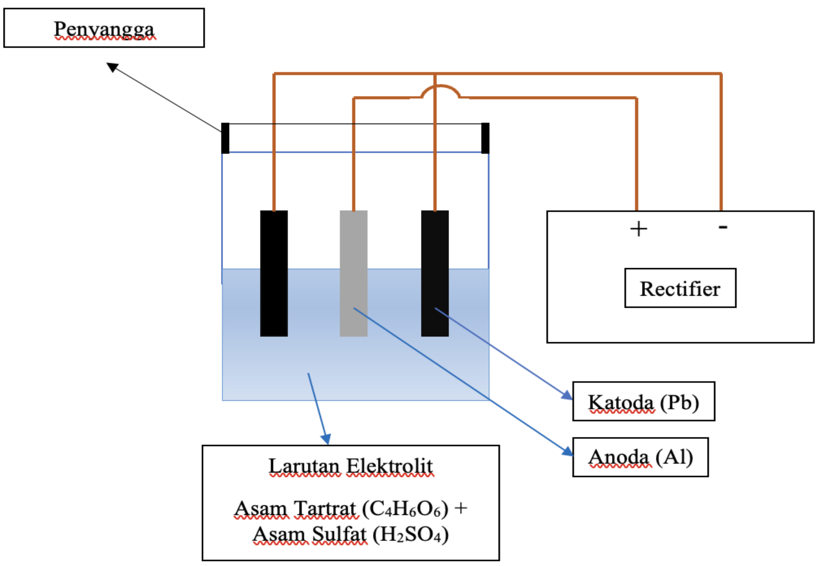
Making the specimens used in this research consisted of the weighing process, measuring 30x80x0.25mm specimens, cutting the specimens for initial surface preparation, and the hard anodizing process. The material used is Al series 1XXX, totaling 9 pieces. To ensure what elements are contained in the material, spectrometric testing is carried out, then the results of the spectro testing will be adjusted to AL standards to ensure details of the material used.



Picture 1. Specimen

2. Hard anodizing process

Al which has a thin oxide layer will increase in thickness if anodizing is carried out. In general, anodizing is carried out at room temperature and for aesthetic purposes, while hard anodizing is carried out at low temperatures and tends to be for technical applications. Hard anodizing is carried out with a solution composition of H_2SO_4 (sulfuric acid) 45 g/L and variations in the addition of $C_4H_6O_6$ (tartaric acid) 70, 75, and 80 g/L and with varying times of 60, 120 and 180 minutes . All processes were carried out at room temperature ±25°C.



Picture 2. Anodizing Scheme

Expected results after doing it. Hard anodizing is an increase in the thickness of the oxide layer on the surface of the material and a porous layer is formed with a micro size (Nugroho, 2015) . The thickness of the layer formed will be proven through microscopic measurements using metallographic testing. Coating elements and compounds will be proven through EDS and XRD testing. To determine the surface morphology and porous formed, SEM testing was carried out with a magnification of 5000x to 10,000x.

Results and Discussion

A. Chemical composition

In this research, material with a high purity value will help the anodizing process achieve results that meet the criteria. Based on this, here are several references that also explain things to ensure optimal growth of the oxide layer. References: (Ward et al., 2003) , (Schwirn et al., 2008) , (Kikuchi et al., 2020) , (Bruera et al., 2020) .

Table 1
Composition of Material Elements in Spectrophotometry Testing.

No.	Elements	Concentration (%)	
		Specimen	Aluminum Association 1100
1.	Aluminum (Al)	99.00	Balance
2.	Iron (Fe)	0.41	0.95
3.	Magnesium (Mg)	0.20	-
4.	Zinc (Zn)	0.1	0.1
6.	Copper (Cu)	0.04	0.05-0.2
7.	Titanium (Ti)	0.02	-
8.	Lead (Pb)	0.01	-

Based on the test results, it can be seen that the aluminum concentration in the Spectrophotometry test is 99%, so the material used is 1xxx series aluminum. According to the Aluminum Association, the specimen most closely approximates 1100 series aluminum (Bisioni et al., 2019) .

B. Metallography

The purpose of metallographic examination is to compare Al with layers and determine the thickness of the layer resulting from the hard anodizing process. Testing was carried out using qualitative metallographic methods, namely using an optical microscope. The etching solution used in metallographic examination is HNO_3 dissolved in distilled water. All specimens to be examined will be focused at 1,000x magnification. The results of the metallography examination can be seen in figures 1 to 5.
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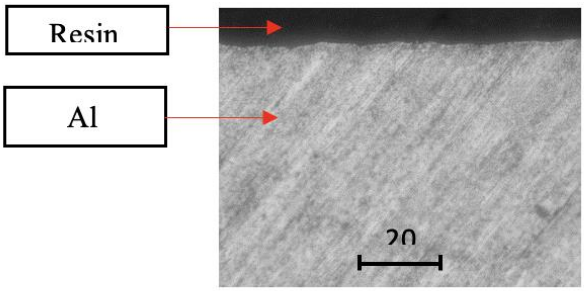


Figure 3. Metallographic Test Results Before Hard Anodizing Process.

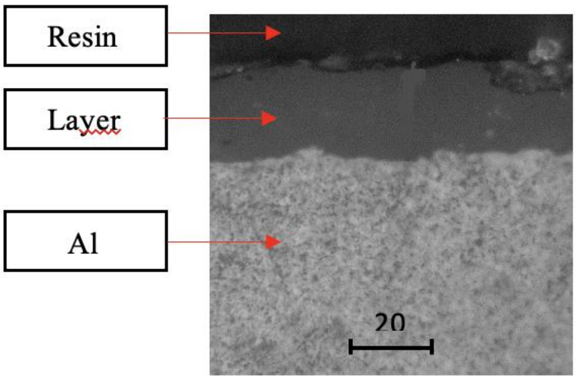


Figure 4. Metallographic Test Results after Hard Anodizing Process

The results of metallographic testing with 1,000x magnification show that Al was before the hard anodizing process, as shown in Figure 1. This image shows that the specimen has no layers, only Al and resin are visible. Meanwhile, the specimen after the hard anodizing process, as shown in Figure 2 with 1,000x magnification, has a clearly visible gray layer. It can be seen that the specimen is white, the resin is black and the coating is gray.

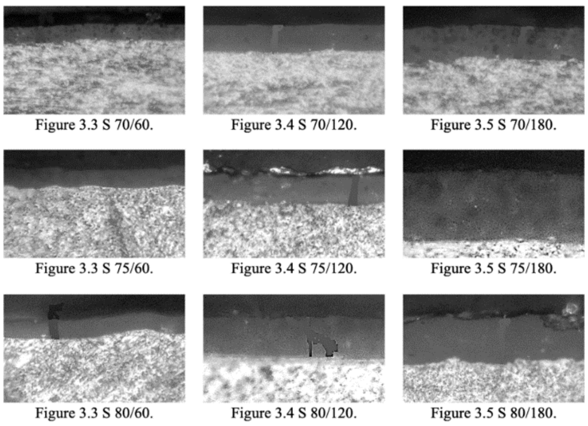


Figure 5. Metallography Results

In the metallography data, it can be seen that the Al alloy that undergoes the hard anodizing process will have a thicker layer. On the other hand, Al that has not undergone this process does not appear to have layers, even though there is actually a thin Al_2O_3

layer that is not visible when tested at 1,000x magnification. The results of metallographic tests on specimens undergoing the hard anodizing process show that the aluminum part appears bright white, while the layer formed appears dark gray on the outside of the aluminum plate, and the resin appears black.

C. Layer Thickness Value

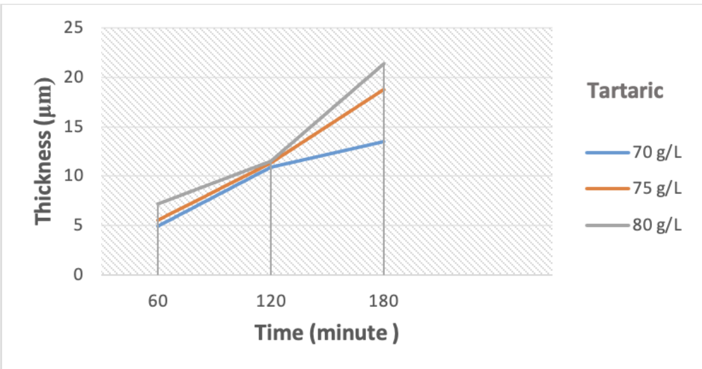
Tell me the amperes and voltage

Measurement of the thickness of the oxide layer was carried out to determine the growth of the oxide layer formed on the 1xxx series Aluminum Alloy after going through a room temperature hard anodizing process and a solution variation of 45 g/L H_2SO_4 and 70, 75, 80 g/L $C_4H_6O_6$ are shown in the following table:

Table 2
Table of Results of Inspection of Layer Thickness Value After Hard Anodizing Process.

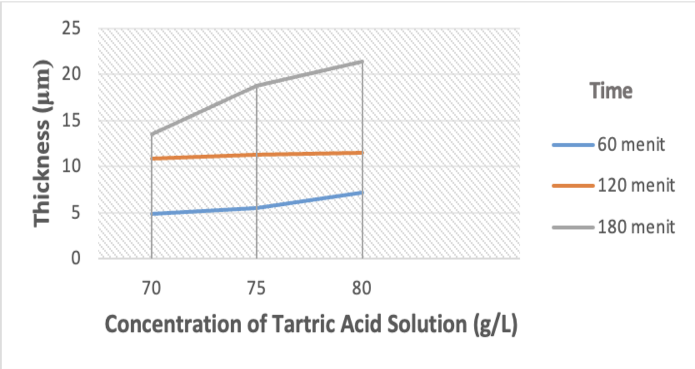
SP	70	75	80
60	4,901	5,509	7,196
120	10.87	11.30	11.51
180	13.47	18.72	21.37

Based on the results of thickness measurements using the metallographic testing method, it can be seen that Al alloys that go through the hard anodizing process have variations in layer thickness. The data obtained was converted into a graph showing the comparison of layer thickness with the time of the hard anodizing process used, which can be seen in Figure 6. Another graph showing the effect of adding tartaric acid on layer thickness is shown in Figure 6.



Picture 6. Graph of Hard Anodizing Time against Layer Thickness

Picture 7
Graph of Tartric Acid Concentration against Layer Thickness



The results of measuring the thickness of the oxide layer on 9 specimens show the development of thickness after the hard anodizing process. The phenomenon of increasing layer thickness runs normally, with time variations of 60, 120, and 180 minutes and a solution variation of 45 g/L H_2SO_4 and 70, 75, and 80 g/L $C_4H_6O_6$ affect the resulting data. The minimum thickness value was obtained at 60 minutes and a combination of 45 g/L $H_2SO_{4\ \mathrm{solution}}$ and 70 g/L $C_4H_6O_6$, with a yield of 4.901 µm. Meanwhile, the maximum thickness value was reached at 180 minutes and a solution combination of 45 g/L H_2SO_4 and 80 g/L $C_4H_6O_6$, which is 21.37 µm.

D. X-Ray Diffraction (XRD) Specimens from Hard Anodizing Process

The purpose of the X-Ray Diffraction (XRD) test is to determine the phases and compounds that form on the surface of the specimen. The results of the X-Ray Diffraction (XRD) test can be found in table 3 and figure 8.

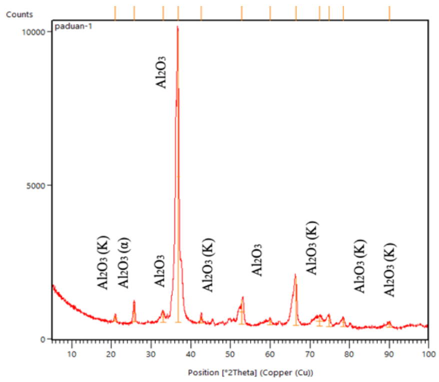


Figure 8. XRD Testing Graph.

Based on the X-Ray Diffraction (XRD) test data in table 3 and figure 8 shows information about the phases and compounds formed on the surface of the specimen. The results can be seen in tables and figures. The phase identified on the surface of the specimen is α and the compound formed is Al_2O_3.

E. Scanning Electron Microscope (SEM)

Scanning Electron Microscopy (SEM) was carried out to observe the microstructure of aluminum alloy which underwent a hard anodizing process at room temperature with a solution composition of 45 g/L H_2SO_4 and 80 g/L $C_4H_6O_6$ and hard anodizing duration of 180 minutes. This study focuses on the surface of the specimen, the results can be found in figures 9 to 13 and table 4 regarding the size of the pores formed.

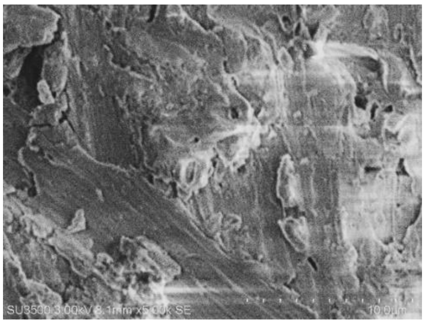
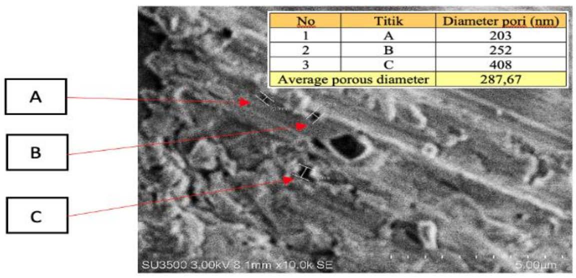


Figure 9. SEM Test Results Magnification of 5,000



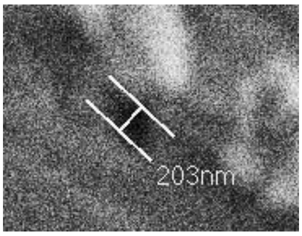
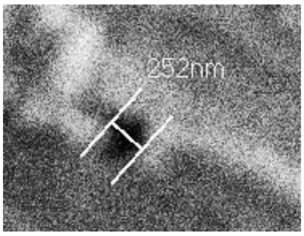


Figure 11 Point A



Picture 12 Point B

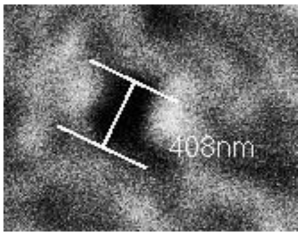


Figure 13 Point C

Figure 10. SEM Test Results from 10,000 X Magnification SP 80/180.

SEM test results with magnifications of 5,000 and 10,000 X show differences in data compared to existing references. This shows that the SEM testing carried out was not optimal. Generally, to see the diameter of the Al_2O_3 layer formed, a minimum magnification of 50,000x is required. However, in this study only 10,000x magnification was used to measure the porous diameter, so the measurement results were less than optimal because only large porous could be measured.

According to literature studies from (Fournelle et al., 2020) and (Aghili et al., 2019) , the porous formed on both sides will be effective if used as a fat filter. It is known that the porous formed with tartaric acid has a diameter of 20 nm which is smaller than that of oxalic acid which produces a diameter of 80 nm.

The SEM test results with 10,000x magnification in Figure 4.15 to Figure 19 and Table 4 show that there is porousness on the surface of the specimen. The measured porous diameter ranged from 203-408 nm. This diameter was obtained from experiments with a 45 g/L $H_2SO_{4\,solution}$ and 75 g/L$C_4H_6O_6$ with a hard anodizing time of 180 minutes.

Although the variations used in the hard anodizing process have not produced porous material that penetrates the thickness of the specimen, this experiment is in accordance with Moesey Lerner's theory. However, for filter applications, variations in solution and hard anodizing time are not optimal because the porous formed is not able to penetrate the thick layer of the specimen.

F. Electron Dispersive Spectrum (EDS) Examination

EDS testing was carried out on the S80/180 specimen to study how variations in the time of the hard anodizing process and the addition of tartaric acid to the sulfuric acid electrolyte affected the elements formed after the hard anodizing process at room temperature on two parts of the specimen surface. EDS test results data can be seen in figures 14 to 16 and tables 1 to 3.

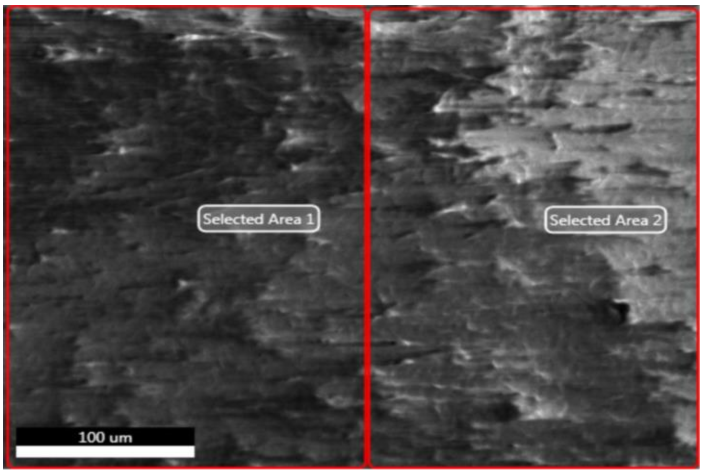


Figure 14. EDS Test Results in Full Area 1 and 2.

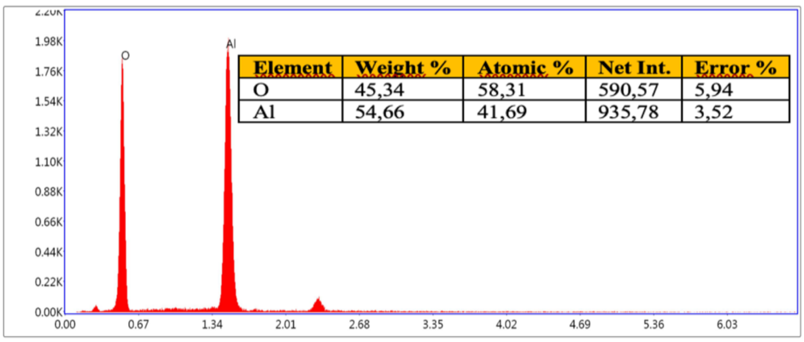


Figure 15. Graph and Composition Value of EDS Full Area Test Results from 1.

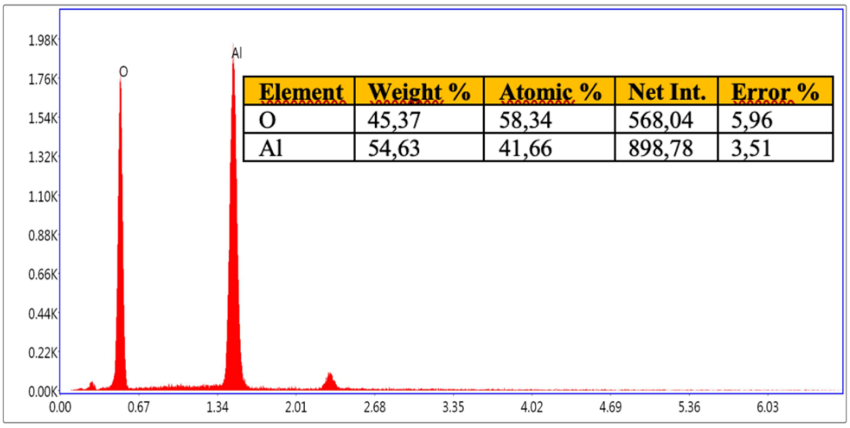


Figure 16. Graph and Composition Value of EDS Full Area Test Results from 2.

Several theories discovered from books and confirmed by journals state that aluminum oxide (Al_2O_3) has hexagonal dimensions and has porosity in the center. Based on this theory, it is believed that the results of the hard anodizing process carried out in this experiment will produce an $Al_2O_{3\ layer}$ which has dimensions according to theory and has porosity.

To prove that the layer formed after the hard anodizing process is truly an $Al_2O_{3\ layer}$, EDS testing can be carried out. EDS test result data can be found in Figures 14 to Figure 16 and Tables 1 to Table 3. From the EDS test results data, it can be seen that the elements contained in the layer formed after the hard anodizing process are Al and O elements. However, to be sure that the layer formed is Al_2O_3 , it needs support from other tests, so XRD testing is carried out .

Conclusion

The conclusions obtained from the discussion are: (a) The aluminum used in this research is AA 1100. (b) The time used is 60, 120 and 180 minutes. This shows that the longer the hard anodizing process takes, the greater the thickness. From a thickness of 4.901 µm in an experiment with a time of 60 minutes to a thickness of 21.37 µm in an experiment with a time of 180 minutes. (c) Addition of $C_4H_6O_6$ 70, 75 and 80 g/L. It shows that the higher the concentration, the addition of $C_4H_6O_6$ results in an increasing thickness. From a thickness of 4.901 µm with the addition of $C_4H_6O_6$ 70 g/L to a thickness of 21.37 µm in the experiment with the addition of $C_4H_6O_6$ 80 g/L. (d) SEM test results on S80/180 show a porous diameter of 203-408 nm, with a hard anodizing process time of 180 minutes and the addition of $C_4H_6O_6$ 80 g/L.

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