The effect of electrolyte concentration and electric current on the quality of surface colouring on anodized aluminium

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Abstract. The use of aluminium metal in daily life has widely used colouring techniques to enhance the aesthetic value of the metal. Aluminium anodizing process can produce porous on the metal surface. The formed porous can be used to store and hold the colouring agent to make them more durable. The research intends to observe the colouring characteristics on the aluminium surface influenced by several parameters of the anodizing process, including electrolyte concentration and electric current. In this study the current H2SO4 concentration was used as a variable to improve the quality of staining on anodised aluminium surfaces. The anodization process was carried out on H2SO4 electrolyte solution with variations in concentrations of 10% to 20% and the current density used was 3 A to 5 A. The tests were carried out using micro Vickers to observe the hardness value. The hardness was higher at lower concentration of electrolyte solution due to thinner layer of oxide film, scanning electron microscope to observe the structure and visual observations for anodised colour quality. From the results given, the tendency of the lower current density the size and density of porous lower. With the same condition, the colour was darker than higher current density.

1. Introduction
Aluminium has a unique and interesting characteristic of natural white colour. The addition of colour to the metal surface enhances the aesthetic value of aluminium products. Aluminium using colouring is often found in components in the automotive industry, household goods and so on. In the future, the need for aluminium in the world is predicted to increase due to environmental issues, population growth, urbanization and technological improvements and innovation. So that the development of technology related to aluminium is needed.

Naturally, the surface of aluminium will produce aluminium oxide layer which makes aluminium metal more resistant to the environment better known as corrosion reaction. Anodization is an electrolysis process that is carried out to produce an oxide layer that is thicker than the naturally formed oxide layer. Resistance to corrosion in the environment will be obtained if the anodizing process is successfully carried out appropriately. In general, anodizing is the process of converting coatings on aluminium metal surfaces and their alloys to become porous aluminium oxide (Al2O3) layers. The overall reaction that takes place during anodization is:

\[ 2\text{Al} + 3\text{H}_2\text{O} \rightarrow \text{Al}_2\text{O}_3 + 3\text{H}_2 \]  

(1)

The reactions at the anode occur at the metal/oxide, can be seen as follows:

\[ 2\text{Al} + 3\text{O}^2- \rightarrow \text{Al}_2\text{O}_3 + 6\text{e}^- \]  

(2)

The reaction at the cathode is hydrogen gas evolution:

\[ 6\text{H}^+ + 6\text{e}^- \rightarrow 3\text{H}_2 \]  

(3)
Basically, anodizing process is very similar to electroplating process. Aluminium metal is coated, but in this case the aluminium is covered in a porous aluminium oxide. The coating process also uses electrolyte solutions. The distinguishes of the two is, in the anodization process, the metal to be coated is placed at the anode [1]. Electrolyte solutions used commonly are acidic[2]. The layer formed will depend on the process parameters. The reaction that occurs is a liquid-solid reaction in the electrolyte solution. The phase of the oxide layer is amorphous. The composition of the layer includes the thin barrier layer and porous outer layer. The shape of the pores is tubular and regular [3]

With anodizing technology, we are trying to develop a process of colouring the surface of metals, especially aluminium, to be better and more durable. There are several parameters that influence the anodizing process, but in this study, we only use the parameters of the current and electrolyte concentration to produce a better colouring. This research is a series of previous studies conducted by the author[4], which present the characteristics of porous on aluminium surfaces which are influenced by electric current and electrolyte concentration. From the results of these studies indicate that these two parameters will affect the distribution and pore size. Besides that, the characteristics of the porous also affect the hardness of the aluminium surface which has been anodized. The further development is to add colour onto anodized aluminium surface. The purpose of this study is to find the optimum conditions for the aluminium surface colouring process with aluminium oxide coating media as a binding agent for dyes

2. Experimental method
In this study using aluminium of the Al100 series which is containing Aluminium up to 99% wt. Then aluminium is attached to an electrical circuit as seen at figure 1. Aluminium is placed on the anode side while lead is placed on the cathode side. This experiment uses electrolyte solution H₂SO₄ which is diluted with water to 10%, 15% and 20%. Besides being inexpensive and easy to obtain, oxide layer results using sulfuric acid electrolytes have extensive aesthetic and functional properties. The circuit is connected to a direct current (DC) source set at 3 A, 4 A and 5 A of electric current as seen in table 1. After lead and aluminium metals are attached to each pole, then dipped in electrolyte solution. The duration of immersion is 60 minutes at 25 V of voltages. Anodizing reaction is considered ongoing if there are bubbles appear on the surface of the bath. After the anodization process is complete, all workpieces are cleaned with water, then followed by the colouring process.

![Scheme of the anodization process](image)

Figure 1. Scheme of the anodization process

In the colouring process, anodized aluminium is dipped in a dye solution (15 gr / litter) which is added to the water for 10 seconds. This colouring process is not only serving to provide colour to add decorative value to aluminium metal, but also as a protective layer on the oxide layer. After the colouring process, then continued with a sealing process which aims to close the porous of the oxide layer formed in the anodizing process, besides the function of this process as a colour lock. In this process use vinegar acid solution (20 ml / litter) of water, with the duration of immersion for about 10 seconds.
Table 1. Experimental variables

| Sample Code | Electric Current (A) | [H₂SO₄] % | Holding Time (minute) | Voltage (V) |
|-------------|---------------------|-----------|-----------------------|-------------|
| A           | 3 A                 | 10        |                       |             |
| B           | 4 A                 | 10        |                       |             |
| C           | 5 A                 | 10        |                       |             |
| D           | 3 A                 | 15        |                       |             |
| E           | 4 A                 | 15        | 60                    | 25 V        |
| F           | 5 A                 | 15        |                       |             |
| G           | 3 A                 | 20        |                       |             |
| H           | 4 A                 | 20        |                       |             |
| I           | 5 A                 | 20        |                       |             |

After a series of anodizing, colouring and sealing processes have been completed, the next process will be carried out by testing and observing the product from the anodizing process. In this experiment, tests and observations made on specimens included visual testing to see the colour quality of the product, measurement of the size and density of porous on the surface using an optical microscope, then the thickness of the oxide layer using a scanning electron microscope.

3. Result dan Discussion
The anodizing process produces porous on the aluminium metal surface as shown in Figure 2. The treatment of each parameter causes the size and distribution of porous to be different. The image on the figure 2.a. has lower electric current and concentration of electrolyte solution which has the smallest average pore, while on the right on the figure b, the electric current and electrolyte concentration is higher.

![Figure 2. Porous appearance on Aluminium anodizing surface.](image)

3.1 Characteristics of porous
The shape of the porous produced on the surface of the aluminum metal is round. The value of electric current and electrolyte solution affect to the size of the average pore diameter. The smallest diameter size is obtained from the condition of 3 A current and 10% electrolyte concentration, which is 17.53 μm, and the highest in the condition of 5 A current and 15% electrolyte concentration, which is 29.18 μm. As seen in figure 3. In this study the formed porous does not look distributed evenly on the surface of the aluminum surface. This could have happened because the anodizing time was too short while the time of the experiment carried out by other researchers was around 20 hours [5].
3.2 Density of porous
The density of the porous depend on the distribution of the pores. The density will raise with the increasing electric current. But different things are shown to variable of the electrolyte concentration. The higher density of pores value is not followed by higher concentration of electrolyte (figure 4).

3.3 Characteristic of color
Visual observations of the anodized aluminum plate surface were carried out. Code samples A and B show anodized results with 3 A and 4 A currents while the electrolyte concentration is 10%. The color shown is denser and darker than the sample with the others (figure 5). This could be caused by thicker oxide thickness in specimens A which reached 103 mm as seen in figure 6.a, compared to H specimens.
which only reached 18.67 mm in thickness (figure 6.b). This possibility is the cause of hardness difference each of the specimens.

![Figure 5. Characteristic of color on Aluminum Surface per experiment sample](image)

A is higher than specimen H, up to 59.1 VHN while specimen H only up to 51.2 VHN. After coloring process, the hardness value increases a little. Higher electric current makes the color on the plate surface is more distribute not evenly.
Figure 6. The thickness of oxide layer (dark grey) on the top of base metal (light colour) on Specimen A (a) and Specimen H (b)

4. Conclusion

Process parameters such as electrolyte concentration and electric current can produce different oxide layers characteristic on the surface of aluminium metal. The oxide layer is in the form of a pore which is expected to store the dyes in it, but the resistance of colour at the certain time on the surface still need to be explored more for further research especially in the interaction of the process. However, in this study, the pore characteristics can affect the colour density on the anodised aluminium surface.

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