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Effect of Current Density Distribution on The Coating Formation in Anodization of Aluminum Using Multiphysics Simulation

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Abstract— **Distribution of electric field in the electrolyser during aluminium anodizing is one of several crucial factors, affecting formation of coatings on the aluminum substrates. In this study, a relatively simple COMSOL model has considered to assist the observation of the distributions of the current density under different types of electric field that determined by different distance between the working electrode (anode) and the cathode. The approach of the study attempts by combining the simulation results with some literature experiments influences in current distribution during anodization of aluminum.**

In order to investigate the effects of the distance between the working electrodes inside the cell on the growth rate anodized film, five different distances between the anode and cathode have been proposed in this study. The simulation results indicate that the current distributes non-uniformly along the substrate surfaces and decreases from the edges to the center. With increasing distance between the anode and cathode, the current density from edges to the center get smaller. The simulation results showed also, that the electrode distance strongly influences the current density due to the current field distribution in the electrolyser. In all cases, the current density values at the edges are approximately twice the values of on the middle.

Index Terms: **anodizing, current distribution, simulation, distance, thickness.**

I. INTRODUCTION

Anodization is a synthesis technique where a protective oxide layer is formed on the surface of metal substrate by electrolytic oxidation of metal surface in the presence of an acid solution. Morphology and thickness of aluminum anodizing coatings are important at all applications of protective finish to ensure optimum performance is obtained [1, 2]. Coating thickness uniformity is one of the important factors influencing the performance of anodizing coatings on aluminum alloys.

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It affects the overall performance of the coating, which's not only helps make that good first impression but the overall evaluation of the product throughout its life cycle [3, 4]. Under the current technological conditions, the primary objective in the field of surface treatment is to achieve a "functional surface". Because of its considerable weight and cost, the construction parts of machinery or building components are designed with relatively fewer reserves, so materials are burdened up to the limits of their possible properties [5].

In anodizing process, parameters such as current and voltage modes, metal substrate, and process time were studied as well as their impact on coating morphologies and properties [4, 6, 7]. However rare studies paid attention to the influence of current density along the working electrode. The current distribution is among the most significant parameters characterizing the operation of the electrochemical cell. The current density on the electrodes is directly proportional to the reaction rate and its distribution critically affects the electrochemical process [8]. In electrochemical cell design, you need to consider three current distribution classes in the electrolyte and electrodes.

These are called primary, secondary, and tertiary, and refer to different approximations that apply depending on the relative significance of solution resistance, finite electrode kinetics, and mass transport. Electrochemical cells are known to be characterised by the relation of the current cell passes to the voltage across it, and the current density may not be uniformly distributed on the working electrode surface during anodizing process which leads to poor performance due to uneven coating thickness and low degree of coatings uniformity [4, 9, 10]. The current distribution depends on several factors such as, cell geometry, cell operating conditions, electrolyte conductivity, electrode kinetics, mass transport of the reactants and mass transport of ions in the electrolyte [11]. Because of this complexity, many applications benefit from suitable simplification when modeling. If one of these factors dominates the cell behavior, the others may not need to be taken into account. Therefore, successive approximations are introduced by the classifications of primary, secondary, and tertiary current distribution. Jun Lee and Jong Kim [12] when discussing the effect of current density on porous film formation in anodizing for Al alloy stated that the Pore diameter and interpore distance tended to increase in proportion to the increase in the applied current density. Also, Xun Ma [13] in discussion of the influence of electrode distance between anode and cathode during high voltage anodizing stated the surface morphology and thickness of the coatings are influenced by the electrode distance, although there is no influence on the phase composition. However, the coating on the front side of the sample is thicker than that on the back side of the same sample. Melhem et al. [14] suggested that distance can play a role, depending on the process conditions, and should nonetheless be taken into account. Also, Wei et al. [15] demonstrated that the distance between anode and cathode affected the anodic current and the oxidation efficiency. Therefore, the distance effect has to be considered.

Numerical simulation provides a capability for evaluation of the current distribution in a particular configuration of electrolyser and coating conditions [8]. Since experiments do not offer direct access to the current distribution in anodizing process, especially at the sample surface, the approach of this study can be accomplished numerically by modeling and simulation. Thus, the objective of this study is to investigate the effects of the distance between the working electrode and the cathode inside the cell on characteristics of current density during the anodizing process by develop a modelling methodology which considers current density-sample length diagram.

II. MODELING AND SIMULATION

To achieve better insights into the current density distribution along the surfaces of the working electrode during the anodizing process and corresponding effects on the coating formation, a numerical model was developed based on a COMSOL-5.6 Multiphysics software [16]. The COMSOL multiphysics commercial software package is used for electrical characterization of the current density distribution in this study. In order to investigate effects of the distance between the working electrode (anode) and the cathode inside the cell on characteristics of current density during the anodizing process and resulting coatings, five different distances between the working electrode and the cathode have been proposed in this study. The obtained simulated results were compared with some experimental results from different literatures. The cell configuration is composed of "working electrode" (anode) faced by the "counter electrode" (tank as cathode) which is surrounds the anode in an even circular shape. Figure 1 shows a systematic 2D geometry of electrodes layout for the electric field analysis samples model and the distances between the electrodes.

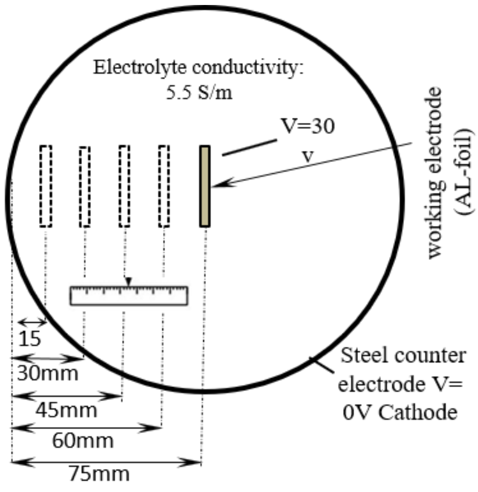

Figure 1. View of the 2 D. model domain and boundaries representing the sample placed in different distances form the counter electrode in the electrolyte.

According to the electrochemical principles process [8, 10, 17], two-dimensional numerical model used in this study is incorporated from the AC/DC module with the following built-in equations:

$$J = -\sigma . \nabla\varphi \quad (1)$$

Where the current density j at any point in the electrolyte depends on the gradient of local potential φ and σ is the electrolyte conductivity.

$$\nabla . E = \frac{\rho}{\varepsilon} = 0 \quad (2)$$

where ∇ · E is the divergence of the electric field, ε is the dielectric constant, and in the electrolyte ρ is the total electric charge density exchange which is 0 (current conservation). So, the potential distribution in the electrolyte can be described by the Laplace equation.

$$\nabla^2 . \varphi = 0 \quad (3)$$

While the boundary conditions for all the insulation walls, there is no current flow.

$$n . J = 0 \quad (4)$$

The model is stationary which means changing conditions with time is not taken into consideration. Ohm's law is used in combination with a charge balance to describe the distribution of current at the electrodes and in the electrolyte. In this work, the value of electrolyte conductivity σ was setup as 5.5 S/m; V was setup in boundary domain; J_e and Q_j were assumed 0 (no external current density and current source). In the boundary domain, the electrolyser was setup as an electric insulator; counter electrodes were setup as V=0; and the working electrode was set as V=30V to simulate a 30V voltage drop between the electrodes. In the point domain, to avoid the point discharge phenomena, all the points were set as 0V. Finally, the overall working electrode area was set as a constant $0.132m^2$. The results of modeling can assist in observation of the current distribution and average current density on the front side of the substrate influenced by changing electrode distances.

Based on the experimental set-up with model domains consisting of boundaries corresponding to the working electrode, counter electrode, electrolyte, and cell walls, a simplified the following assumptions:

a. The electrolyte has considered as a linear homogeneous conductive medium with constant specific conductivity γ.

b. The electrolyte is stirred and its temperature constant.

c. There is no additional external current density or current source.

d. The anode is assumed to have a constant applied voltage.

e. For all the insulation walls, there is no current flow.

Some Physical input parameters used in the numerical simulation are given in Table 1. The most important is model simplification due to system symmetry should be used as much as possible to reduce the simulation run time. There for, the length of sample in the model expressed from 0 to 15 mm which represents from edge to middle of samples. The finite element mesh is shown in figure 2 at the domain area (electrolyte). The mesh consists of 2142 elements. It was generated by using the automatic mesh creator with the option “finer”. At the anode surface, mesh consists of 23 triangular elements and the mesh area 8 mm^2.

Table 1. Show physical Input parameters used in the numerical simulation

Physical parameter	Value
Working electrical conductivity (S/m)	3.77×10^7
Steel conductivity (S/m)	1.4×10^6
Electric potential on anode (V)	30
Electric potential on cathode (V)	0

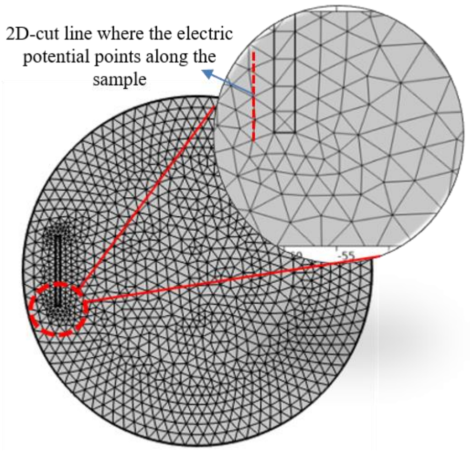

Figure 2. Meshing of the boundaries on the sample (anode)/electrolyte boundary.

III. RESULTS AND DISCUSSION

From the 2D modeling results, figure 3 displays the current density distributions on the surfaces of the front of the working electrodes along its length for different electrode distances between 15 and 75 mm from the cathode (steel tank). The background color and arrows show the distribution and direction of current density.

The difference of the distribution of current density for all cases layout in all electric field conditions can be visualized from the modeling results. Irrespective of the working electrode shape, the electric field strength is the highest near the edges and lowest in the center of the substrate surface for all conditions. This is due to the edge effect [13]. Based on the simulation results which presented in figure 4, the cathode surface area is larger than the working electrode (anode), so the extra cathode exchange current flow into the volume of electrolyte available beyond the anode edge and has to concentrate on the edges. This produces the edge effect and makes the current density at the outside corners higher than that in the middle of the anode surface. The figure shows how the current flows between the electrodes to produce the edge effect. These results consistent with the results reported by C.B Wei *et al* [15, 17, 18].

In order to visualize the simulated distribution of current density (A/m^2) across the sample surface, a 2 D cut line was considered between two points of sample adjacent its front surfaces. Results of calculation the simulated current density distribution along the surface of the working electrode presented in Figure 5. It is observed that the current density values in all cases decreases with an increase in distance between the electrodes (anode and cathode). In all cases, the values of current density at edges are nearly 2 times higher than at the middle. Therefore, disparity values of coatings thickness will be produced on edges and at the middle surfaces. This is also consistent with the results of modelling of electric field and current density distributions on the surfaces of working electrodes discussed in [17].

There are a number of useful review papers which provide an overview of the effects of current density on the surfaces of the working electrodes experimentally.

Hidetaka Asoh et al in [19] investigate the effect of current density and the electrical potential distributions on aluminum at edges and surfaces experimentally. He used aluminum sheets were suspended in an electrolyte solution, and positioned either vertically or horizontally at the center of a cell. He found that the thicknesses of the porous alumina films and barrier layers near the aluminum substrates can be taken to represent the potential distributions. The thickest anodic alumina was formed at the edge of the anodic pole of the bipolar electrode, and its thickness continuously decreased in the direction of the cathodic pole. Experimental results in ref. [13, 20-22] proved that the current flowing through the front and edge surfaces are higher than that flowing through the back and middle surfaces and the average current density decreases with larger electrode distances, which corresponds to the simulation results obtained here. Also C.B. Wei et al stated in discussion of anode current effects in high voltage anodizing that the experimental results demonstrate that the distance between the cathode and anode affects the anode current and the oxidation efficiency. In addition, the current flowing through the front surface (relative to the cathode)

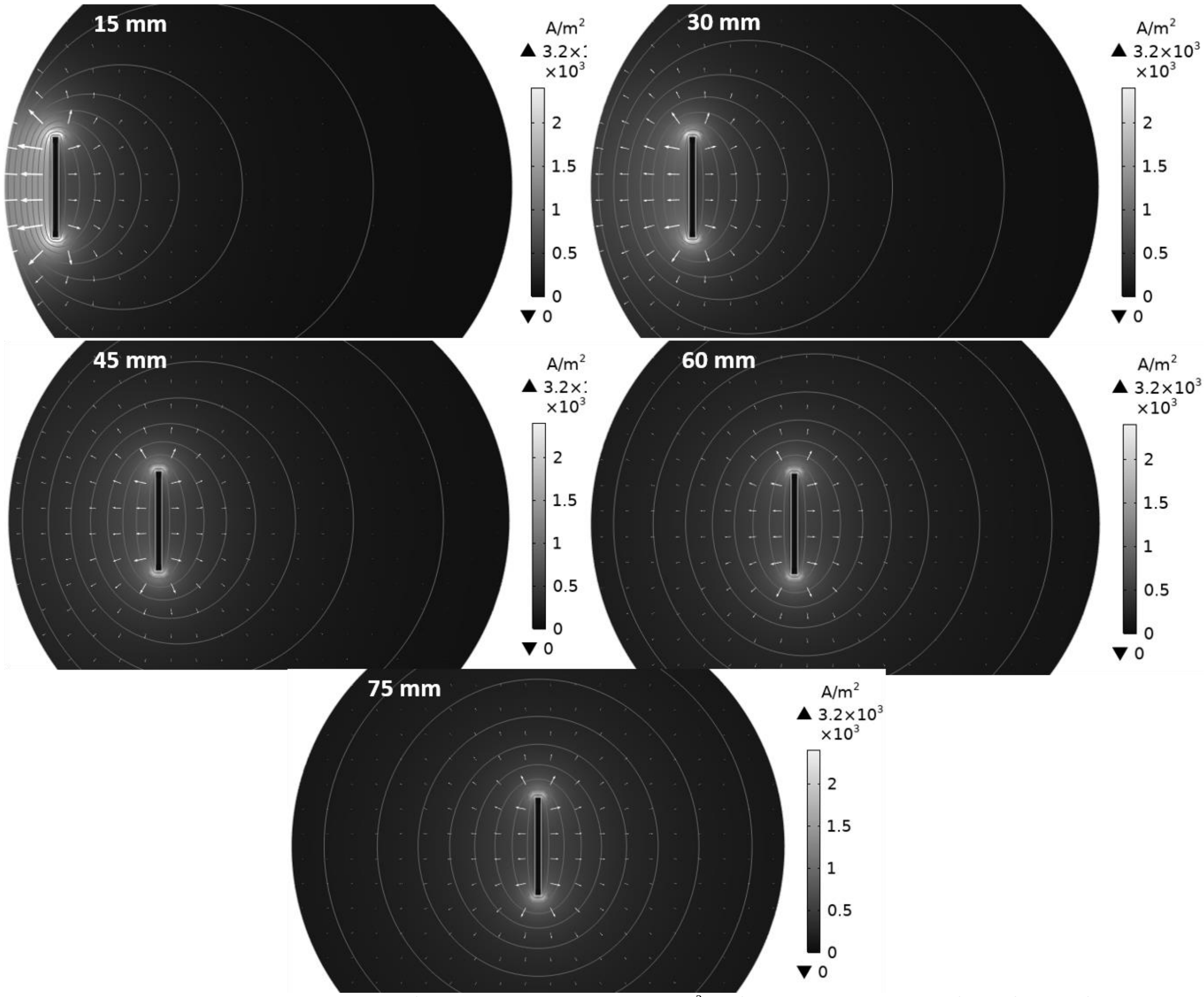

Figure 3. COMSOL modelling results of current density distributions (A/m^2; surface and arrows) on the surfaces of anodes for different electrode distances between 15 and 75 mm.

of the specimen is larger than that flowing through the back surface of the same specimen.

The measured tribological properties and corrosion-resistance agree well with the effects of the current. The front surface exhibits more superior wear and corrosion resistance than the back surface. In other meaning, non-uniform current distribution on the working electrode (anode) leads to thickness variations of anodizing coating on Al substrates. Several authors [4, 22, 23] discussing the importance of the coating thickness in the mechanical properties of anodizing coatings stated that thicker coatings usually show better mechanical properties, however this is not always true as the defects such as porosity may play crucial rule in the mechanical performance of thick anodizing coatings. Design engineers specify thickness for various reasons, including some applications which need specific aim, such as the need of a degree of flexibility which influence by the residual metal in the component. Based on the above data

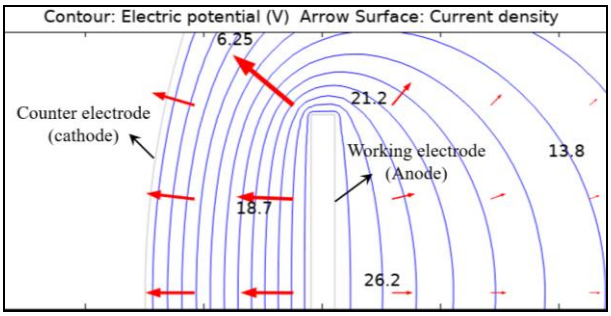

Figure 4. Modelling results of electric potential [V] and current density [A/m^2] distribution respectively as contour and arrow lines

of both simulations and experimental results, it is important to say that the place of counter electrode (cathode) inside the cell affects the deposited coatings are formed and their properties. Also, the thickness of the oxide layer formed. This is the reason why provide better understanding of the electrochemical behaviour and coating formation during the anodizing process.

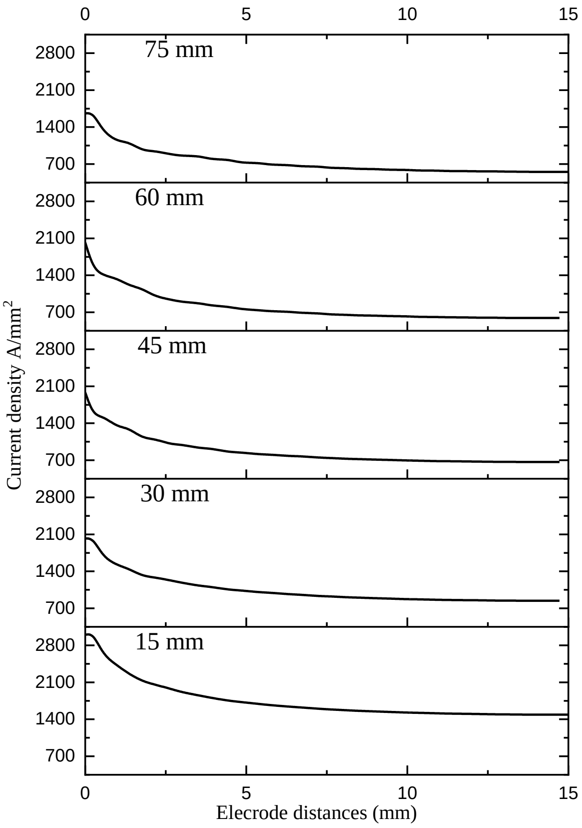

Figure 5. Simulated current density distribution along the surface of the working electrode.

CONLUSIONS

- Numerical values were obtained using a COMSOL Multiphysics package to analyze the magnitudes and directions of the current density in the electrolyser during the anodizing process, taking into account the distances between the electrodes.
- The simulation results indicate that the current distributes non-uniformly along the samples surfaces and decreases from the edges to the center. With increasing distance, the current density from edges to the center get smaller, which means that the current distribution becomes more and more uniform.
- The modelling results showed that the electrode distance strongly influences the current density due to the current field distribution in the electrolyser. In all cases, the values of current density at edges are nearly 2 times higher than at the middle.
- Design engineers can be using the simple modeling approach to optimize the anodizing setup for controlling coating thickness at specific places along sample length.

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