Coating Hydroxyapatite on 316L Stainless Steel Using Electroforesis Deposition Method

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Abstract. Stainless steel 316L is a metal which used as bone plate, however this metal has low biocompatibility in a human body. For increasing their biocompatibility, the stainless steel 316L need to be coated with hydroxyapatite (HA). The purpose of this research is to study the effect of applied voltage and deposition time on the properties of HA coated 316L stainless steel. Stainless steel 316L was cut into 20x20x1 mm³ dimension then HA solution was prepared in ethanol solution used magnetic stirrer for 2 hours and 200 rpm. The HA powder was deposited by electrophoretic deposition (EPD) using carbon as anode and stainless steel 316L as cathode. Powder was coated on stainless steel substrate at 40, 50 and 60 volt and deposition time 10, 20, and 30 minutes. Thickness of HA layer on substrate increased, as the time deposition increased with result of 35 µm, 50 µm and 60 µm at deposition 50 volt and deposition time of 10, 20, and 30 minutes. Thickness HA coatings can decrease the corrosion of stainless steel, in deposition time 10, 20, and 30 minutes is 0.91, 0.704, and 0.56 mpy. at applied voltage 50 volt and deposition time 30 minutes was obtained suitable for bone plate.

1. Introduction
Hydroxyapatite has been known as a good substitute material for bone and dental implants in the world of health due to the similarity of chemical and biological properties with human bone tissue. However hydroxyapatite has poor mechanical strength so it is less suitable to be used as a substitute for bones that support heavy body parts [3].

Implantation is one method of replacing damaged bones and one technique that can be used is bone plate. This bone plate is a component that is used as a recovery process for cracked or broken bones. This technique uses metal as an implant substrate that continues to be developed in accordance with the increasing need for artificial bone in humans. Metals commonly used as bone plates include stainless steel (SS), Cr-Co alloys and Ti alloys [4].

However, the use of metals becomes a constraint in terms of biocompatibility with their hosts (interacting media), including local corrosion, cannot regenerate new bone, limit organ function, and affect bioactivity in the body. In addition, corrosion products will interact with the body and cause implant failure. One effort to improve the biocompatible, fixation and life time of implant material is by coating the metal surface by using certain biomaterials. Bone formation and cell development can be triggered by the modification of coatings with biomaterials or biocramics including hydroxyapatite [4].

Hydroxyapatite with the chemical formula $[\text{Ca}_{10} (\text{PO}_4)_{6} (\text{OH})_2]$ is the majority component that forms human bones and teeth [5]. Therefore, hydroxyapatite has been used for biomedical implants, hard
tissue surgery, bone regeneration, repair, filling, expanding and reconstructing damaged bone tissue [6].

Based on what has been explained above, this idea has the potential to be developed because it can increase the use of metal material as a substitute for damaged bones with better biocompatibility, fixation, and life time by coating hydroxyapatite on a metal surface. In addition this idea can also increase the use of hydroxyapatite to be more maximum.

2. Methodology

The materials used in this study were commercial hydroxyapatite powder, aquades and ethanol as a solvent for the electrophoresis deposition process. While the equipment used in this study is a furnace that serves as a place for the calcination of the product, a magnetic stirrer as a mixer, chemical beaker, measuring cup, porcelain cup as a container for sample, stainless steel metal, carbon electrodes, power supply for electrophoresis deposition process. The HA coating process on the surface of a substrate was carried out 3 steps, namely the preparation of the material, the coating with the EPD technique, and the heating process.

2.1 Raw Material Preparation Process

Before the coating process, the surface of stainless steel metal has been cleaned by grid-500 sandpaper followed by grid-1000 until the surface looks clean, then soaked with ethanol and rinsed with distilled water. Any dirt that might be presented on the metal surface was assume completely clean. 0.5 g hydroxyapatite and 25 mL ethanol was mixed to made hydroxyapatite suspension. After that, the mixture was stirred by a magnetic stirrer with a speed of 200 rpm for 2 hours so that hydroxyapatite could be dispersed in an ethanol solution which was indicated by the turbidity of the suspension by the particles.

2.2 Hydroxyapatite coating on metal surfaces

Hydroxyapatite suspension that has been prepared was inserted into a beaker. Then this solution was connected to the two electrodes, one electrode was a stainless steel metal which was the target of hydroxyapatite coating on its surface (negatively charged) and the other electrode was a carbon plate (positively charged). In the electrophoresis deposition process, hydroxyapatite powder dispersed in a solution would be drove by electrically charged electrophoresis. The hydroxyapatite that was carried would stick to the negatively charged metal surface by forming deposition on the metal surface [2]. Figure 1 shows a series of EPD tools.

![Electrophoretic deposition device circuit](image)

**Figure 1.** Electrophoretic deposition device circuit

2.3 Calcination

After the coating process was completed then the calcination process was carried out at 800 ° C for 2 hours in a furnace.
3. Result and discussion
The characterization of hydroxyapatite that coated on stainless steel metal surface was carried out using SEM, XRD, and corrosion test.

3.1 XRD Analysis
In this study, testing of metal that coated HA with using XRD with voltage variations used is 40, 50, and 60 volts (Figure 2).

![XRD diffractogram](image)

**Figure 2.** Diffractogram of 316L stainless steel coating with HA at 30 minutes deposition time and coating time (a) 60 V (b) 50 V (c) 40 V.

Different peak intensities on HA show different amounts of HA, EPD at 50 voltage results in coatings with more mass or more thickness (dense), which is indicated by higher peak intensities. From the data in Figure 2 it can be obtained that the crests begin to narrow which indicates the crystallinity of the sample begins to rise with the value that can be seen in Table 1.

| Voltage | 2Theta | FWHM | Intensity | Gamma | Cos Θ | Crystal | Mean    |
|---------|--------|------|-----------|--------|--------|---------|---------|
| 40      | 82.7503| 0.575| 100       | 1.5406 | 0.750398| 3.213461|         |
| 50      | 45.4355| 0.55 | 55        | 1.5406 | 0.922418| 2.733013| 2.981691|
|         | 65.5676| 0.55 | 47        | 1.5406 | 0.84072 | 2.9986  |         |
| 60      | 32.3077| 0.26 | 100       | 1.5406 | 0.960518| 5.432945|         |
|         | 32.7601| 0.26 | 65        | 1.5406 | 0.959412| 5.558451| 5.25697 |
|         | 33.4308| 0.309| 61        | 1.5406 | 0.957745| 4.779507|         |
|         | 45.0765| 0.486| 100       | 1.5406 | 0.923624| 3.00888 |         |
| 60      | 82.47  | 0.55 | 75        | 1.5406 | 0.752012| 3.352314| 3.18265 |
|         | 65.28  | 0.53 | 45        | 1.5406 | 0.842076| 3.106742|         |

Based on calculations with the Scherrer equation, the crystal sizes for the voltages of 40V, 50V and 60V are 2.9817, 5.256 and 3.1826. This shows that hydroxyapatite can be deposited well at a voltage of 50 V.
Figure 3. Diffractogram 316L stainless steel coating with HA at deposition time of 30 minutes and a voltage of 50 V.

In Figure 3 it can be seen that there is a similarity of peaks formed in the resulting coating (a) with reference to JCPDS 96-900-2217 (b). This shows that the coating with a voltage of 50V and 30 minutes deposition time is hydroxyapatite, so it can be concluded that hydroxyapatite is completely deposited.

3.2 SEM Analysis

In Figure 4 it can be seen that the HA layer increase by increasing time of coating stainless steel 316L metal with HA. Coating for 10 minutes, the thickness of HA obtained is 35 μm. Increasing the coating time at 20 minutes obtained a thickness of 50 μm, and 60 μm at 30 minutes coating time. During the period of EPD, the relationship between deposition mass and time is linier [1].

Figure 4. Stainless steel 316L cross section that has been coated with HA at a voltage of 60 volts and coating time (a) 10 minutes (b) 20 minutes and (c) 30 minutes

This is due to the large number of charged HA particles deposited on metal surfaces with increasing time. Figure 4.1 (a) shows HA not completely covering metal material, whereas in Figure 4.1 (b) metal material is covered by HA. And in Figure 4.1 (c) the metal material is completely covered by HA and HA is formed, the finer and the more porous. In the layer also found no fractures (cracks).
3.3 Corrosion Test
The thickness of the HA coating that coats the metal is one of the important factors in corrosion resistance. The longer the coating time, it will increase the thickness of the HA layer deposited on the metal. In Figure 5, we will get the corrosion rate data at 10, 20 and 30 minutes coating time variations.

Figure 5 shows that stainless steel 316L that coated with HA has a lower corrosion rate than 316L stainless steel without coating. Corrosion rate values obtained are 0.92, 0.704 and 0.56 mpy, the value obtained will decrease by increasing deposition time. The thicker layer will provide metal protection against corrosion. This result corresponds to [7] that said the increase of the coating thickness enhanced the protection on magnesium alloy substrate and the corrosion rate of the metal decreased.

![Figure 5. Corrosion rate values in the time variation of coating 10, 20 and 30 minutes.](image)

4. Conclusion
Coating HA on stainless steel metal is done with variations in the voltage used and the timing of the coating by electrophoresis deposition (EPD) method. The thickness of deposited HA will increase by increasing coating time on stainless steel metals. The coating produced at a voltage of 50 volts and a deposition time of 30 minutes, can be used as a bone implant because hydroxyapatite is completely deposited, increasing coating time reduces the rate of corrosion of stainless steel. So that the hydroxyapatite coating on 316L stainless steel is able to overcome the problem of bone surgery with a simpler method.

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