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Optimization of the Characteristic of Hydroxyapatite Coating on Ti6Al4V Alloy by Electrophoretic Deposition

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ABSTRACT

In this work, hydroxyapatite (HA) Nano-composite was applied as a coating layer on Ti-6Al-4V alloy by electrophoretic deposition and optimized according to the Taguchi approach. The effect of coating parameters, voltage (40, 60, and 80 V), time (2, 5, and 7 min), and concentrations (1, 2, and 3 g/L) was studied to optimize the quality of HA coating. The porosity, thickness, and Nano-roughness of HA coating layers were characterized by optical microscopy and atomic force microscope (AFM). Results disclosed that the amount of porosity in the coatings decreases from 21.52 % to 0 % as the concentration of HA particles, time of deposit, and voltage value increase. The thickness of HA coating increases as the deposit rate rises. The thickness of the coating prepared with conditions (80 V, 3 g/L, and 5min) is shown the higher value which is 18.304 µm with no observed porosity. The roughness of samples was measured only for the covered samples with a lesser amount of porosity. It is found that the highest Nano-roughness was obtained with sample 9 at 80 V, concentration 3 g/L, and time 5 min and it was 9.5263 nm.

Keywords: *Optimization, Hydroxyapatite, Characteristic, Ti6Al4V alloy, Electrophoretic deposition*

1. INTRODUCTION

Ti alloys are utilized broadly as bio-metal for orthopedic applications due to their resistance to corrosion, high strength-to-weight ratio, and biocompatibility [1, 2]. To improve the implant bioactivity and adhesion with human tissue the surface of the implant should be modified [3-5]. Hydroxyapatite (Ca_{10} $(PO_4)_6(OH)_2$) is a ceramic material utilized as an implant coating because of its similarity to bone chemical composition, bioactivity, and biocompatibility [6–8]. Many coating methods are applied to deposit HA, such as plasma spraying [9], sol-gel [10], and electrochemical deposition [11]. The EPD method is less expensive, simple, less time-consuming, and preferred for complex shapes [12-14]. In experimental design, The Taguchi method offers many advantages over the other approach. It allows the determination of optimal process conditions and important variables efficiently, [15]. The experiments can be managed by using the Taguchi method with variations in parameters to minimize costs while achieve optimal results, [16]. Additionally, the Taguchi design of the experiments method enables the identification of effective factors and their levels, [15]. Also, the Taguchi approach optimizes the components' quantities in materials to meet the strength factors effectively, [17]. However, the Taguchi approach stands out for its ability to reduce costs, and efficiency of determining optimal conditions and enhance the accuracy in various applications. M.K.Abbass, et al. (2021) [18] used EPD process to deposit HA on Ti 6Al 4V alloy with different current once alternating current (AC) and then direct current (DC). Their results shown that the thickness and density of HA coating on the samples were higher if the coated utilizing (AC) current in compared with (DC) current. D. Juliadmi, et al. (2017) [19] coated Ti-6Al-4V alloy with hydroxyapatite by (EPD) process using different voltage and time. Their results cleared that the low voltage and short time deposition gives uncovered samples with less coating thickness. M. Farrokhi-Rad and T. Shahrabin (2014) [20] used the EPD method to deposit HA with different suspensions of alcohols, time, and voltage. Their results showed that the faster deposition rate was in the suspension of butanol. In this work, the controlling parameters of the EPD process have been managed by applying the Taguchi method. The morphology of surface, porosity, and thickness of HA coating have been analyzed by the observing of optical microscopy. The importance of this study is obtaining a coating with good characteristics of the coating such as morphology, thickness, and roughness by using the EPD technique.

2. EXPERIMENTAL WORK

2.1. Materials

Ti6Al4V alloy was provided as a 2 mm thick plate by “Baoji Jinshengmetal Material. Co., Ltd”. The chemical composition of the alloy according to the manufacturer is shown in Table 1.

Table 1 Ti6Al4V alloy chemical compositions

Ti	Al	V	Fe	C	N	H	O	Residual elements
Remainder	6.2	4.1	0.14	0.018	0.024	0.005	0.1	≤0.3

Hydroxyapatite (reagent grade, powder, synthetic) (20 nm, purity of 96%, white color with 502.31 molar mass) was purchased from Hualanchem. Co. China. Ethanol absolute (C_2H_5OH) 99.8%. (Honeywell. Germany).

2.2. Preparation of Samples and Solutions

The Ti6Al4V alloy was cut to (20 x 10 x 2) mm and then ground with SiC emery paper using 220, 400, and 600 grit. The cleaning process used ethanol in an ultrasonic bath for 15 min. At last, the samples were dried at 100 ºC inside an oven for 15 min. The aqueous suspension of HA was prepared by using ethanol then HA nanoparticles were added to the ethanol solution. To ensure the stability of the prepared suspension, the suspension pH was adjusted to 4. The beaker containing the suspension was covered with film to prevent evaporation. The suspension was stirred by a magnetic stirrer to deagglomerate the nanoparticles for 1 hour. Then the particles were dispersion by using an ultrasonic device for 30 minutes and the suspensions were again stirred for a few seconds to move the stagnant particles from the bottom before coating.

2.3. Electrophoretic Deposition Cell

The used EPD deposition cell consisted of two electrodes inside a beaker filled with the prepared solution. The used electrodes were cleaned and dried, and the coating was applied by immersing the electrodes (the Ti6Al4V alloy sample was used as the cathode and the anode was 316L stainless steel) inside a beaker filled with solution. The sample (as cathode) is distanced from the anode by 1.5 cm. The EPD system used in this study is shown in Figure 1.

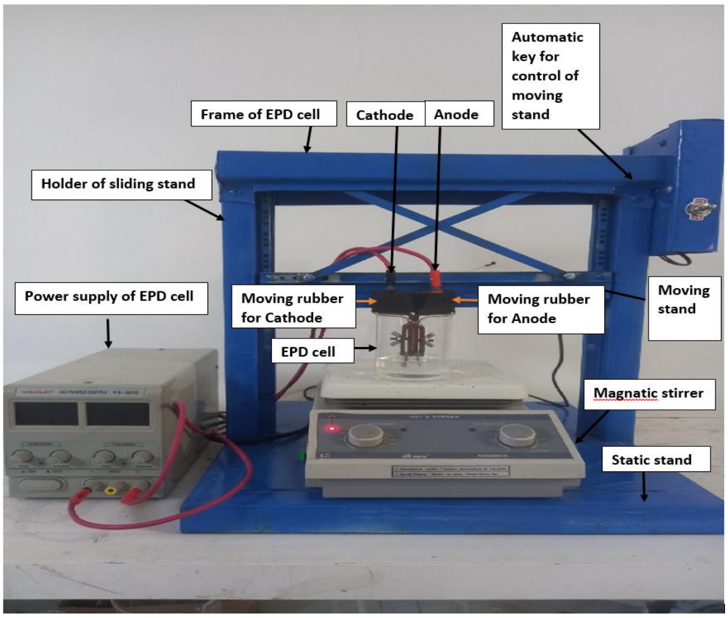

Figure 1. EPD system depositions used.

It is noticed from Figure 1 that the holder of two electrodes is connected to a voltage/constant power source. A moving stand was used to Lifting and lowering the electrodes in the baker which was carried out electrically according to the design of the cell deposition system. The coating was employed during the experiments by applying the selected conditions. The Taguchi method was utilized to choose the optimum parameters of the deposition process. According to the Taguchi method, the array of type (3^3) is shown in Table 2, while Table 3 shows the deposition factors on the chitosan layers.

Table 2 The orthogonal array of L9 (3^3) for deposition of the coating layer

Experimental	Voltage (v)	Concentration (g/L)	Time (min)
1	1	1	1
2	1	2	2
3	1	3	3
4	2	1	2
5	2	2	3
6	2	3	1
7	3	1	3
8	3	2	1
9	3	3	2

Table 3 Deposition factors affect the HA layers

Experimental	Voltage (v)	Concentration (g/L)	Time (min)
1	40	1	2
2	40	2	5
3	40	3	7
4	60	1	5
5	60	2	7
6	60	3	2
7	80	1	7
8	80	2	2
9	80	3	5

2.4. Morphology, Thickness, and Nano-roughness Analyses

The optical microscopy type (MTM-1A) was used for the morphology analysis. For thickness measurement, grounding was used for preparing the cross-sections of the samples with 2000-grit silicon carbide paper. The atomic force microscopy (AFM) technique is an influential surface analysis test used for studying the Nano-surface morphology and roughness of coatings. AFM can analyze the surfaces of materials in an atomic resolution reaching approximately 1 nm or less.

3. RESULTS AND DISCUSSION

3.1. The Porosity of HA Layer

Figure 2 shows images of the different HA coatings obtained with the 9 experiments. Image -J was used to estimate the percentage value of porosity for each sample image in red color and their results in Table 4. As it is expected the amount of porosity in the coatings increases as the concentration of HA particles decreases in the suspension. As can be observed, increasing the value of voltage reduces the amount of porosity notably. The coating prepared with conditions (80 V, 3g/L, and 5min) shows morphology with no amount of porosity. The morphology, and thickness of the coating are affected by the voltage, concentration of powder, and time of deposit. The movement of the particles inside the suspension increases with increasing the potential of current generated by the field of electricity [21]. The rate of deposition increases with increasing the amount of voltage. So, the rate of voltage controls coating quality. High voltage will increase particle velocity which leads to turbulence of suspension. So, the particles with small volume deposit earlier than the particles with large volume [22]. Thus, the arrangement of particles on the surface of samples is ununiformed as the smallest particles agglomerate and cause a high amount of porosity [22-24]. Lowing the amount of voltage slows the movement of particles to a proper level which leads to uniform deposition of the coating. On the other hand, the too-low voltage cannot migrate larger particles due to insufficient energy [25].

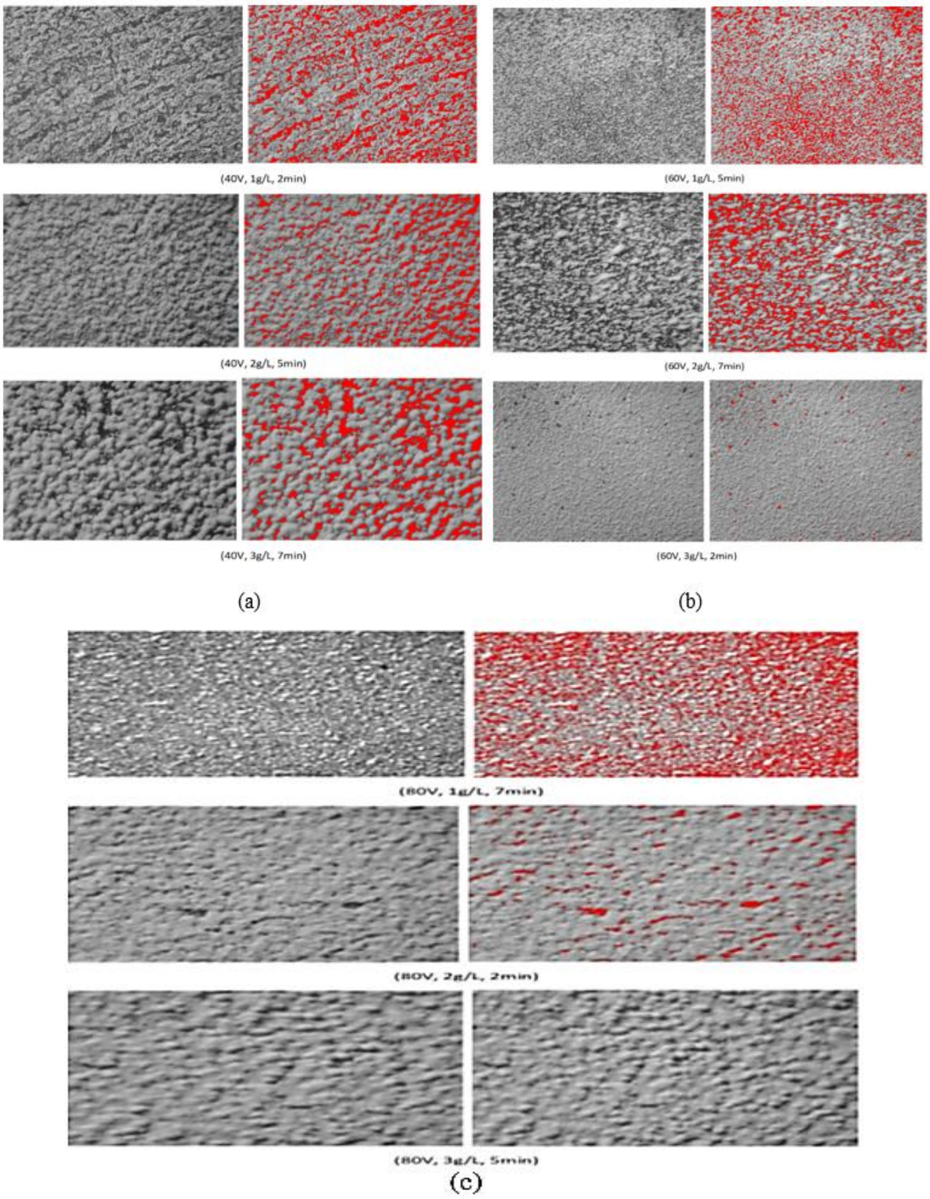

Figure 2. Optical images (at 50 x magnification) of HA layers for (a) experiments 1,2, and 3 (b) experiments 4,5, and 6 (c) experiments 7,8, and 9 prepared according to the Taguchi method.

Table 4 The porosity of HA coating that prepared according to the Taguchi approach

Sample No.	Voltage, V	Concentration, g/L	Time, min	Porosity %
1	40	1	2	21.52
2	40	2	5	15.91
3	40	3	7	17.97
4	60	1	5	20.86
5	60	2	7	21.24
6	60	3	2	0.77
7	80	1	7	25.8
8	80	2	2	3.56
9	80	3	5	0

3.2. The Thickness of the HA Layer

The cross-section of HA coatings obtained for 9 experiments are shown in Figure 2 and Table 5. The thickness of the coating prepared with conditions (80 V, 3g/L, and 5min) is shown the higher value which is 18.304 µm with no porosity.

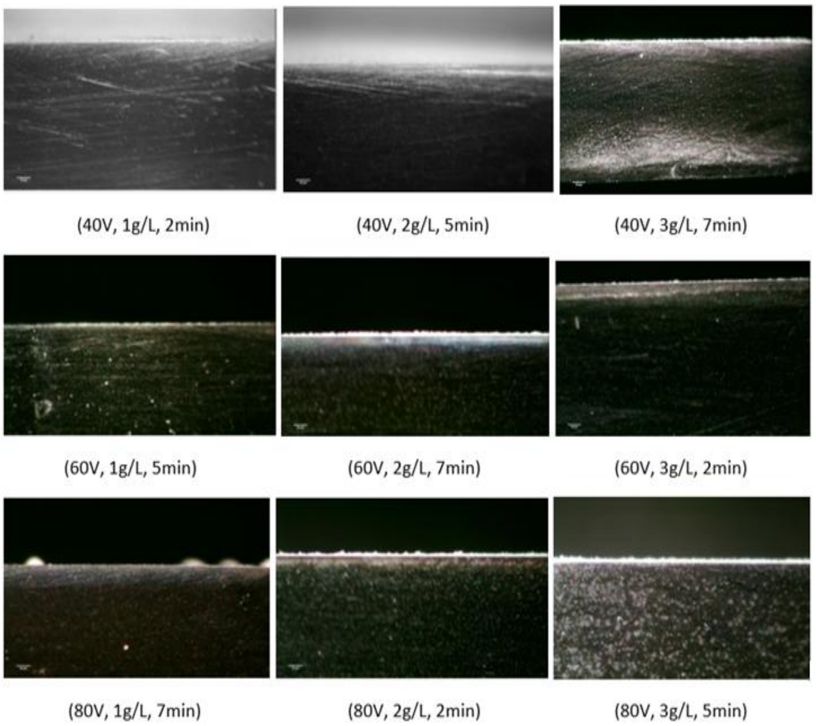

Figure 3. Optical images (at 50 x magnification) for a cross-section of HA coating layers.

Table 5 The thickness of HA coating that prepared according to the Taguchi approach

Sample No.	Voltage, V	Concentration, g/L	Time, min	Thickness (µm)
1	40	1	2	4.242
2	40	2	5	7.655
3	40	3	7	7.615
4	60	1	5	5.775
5	60	2	7	9.88
6	60	3	2	5.896
7	80	1	7	5.452
8	80	2	2	11.111
9	80	3	5	18.304

To select the optimal conditions (voltage, concentration, and time) for depositing HA layer by Taguchi method, S/Ns ratio tables for the thickness (larger-the-better) was utilized to determine the highest thickness of coating. Table 6 reveals S/Ns ratio for the thickness of HA layer depending on the orthogonal array of L9 (3^3).

Table 6 Signal-to-noise ratio of Taguchi design for thickness HA coating layer

Sample No.	Voltage, V	Concentration, g/L	Time, min	Thickness, µm	SNRA1
1	1	1	1	4.242	12.5514
2	1	2	2	7.655	17.6789
3	1	3	3	7.615	17.6334
4	2	1	2	5.775	15.2310
5	2	2	3	9.88	19.8951
6	2	3	1	5.896	15.4111
7	3	1	3	5.452	14.7311
8	3	2	1	11.111	20.9151
9	3	3	2	18.304	25.2509

Table 7 shows the ranking of significance for each level of various factors derived from the obtained data values. The results reveal that the concentration factor has the highest effect on the experiment while the voltage has the second level and the time has the lowest effect.

Table 7 The rank of the controlled factors on the thickness of the HA coating layer

Level	Voltage	Concentration	Time
1	15.95	14.17	16.29
2	16.85	19.50	19.39
3	20.30	19.43	17.42
Delta	4.34	5.33	3.09
Rank	2	1	3

The result of SNs ratio is shown in Figure 3 which clear the way that the factors affecting on the process.

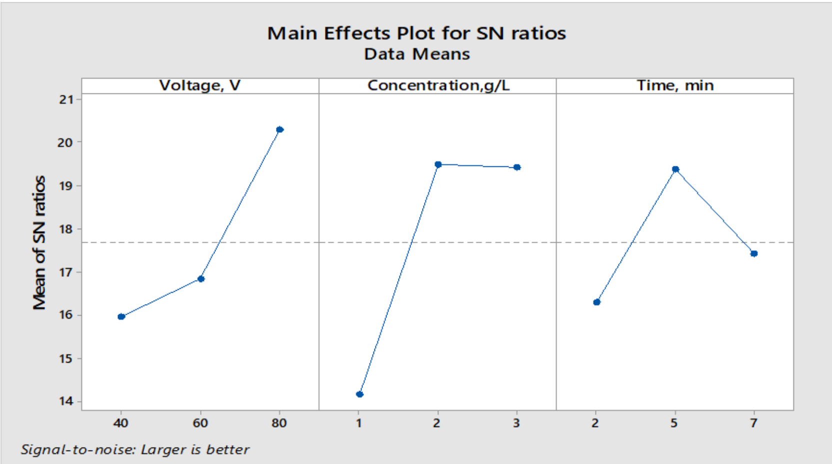

Figure 3. The graph of SNs ratio for thickness of HA coating layer.

The graph of SNs ratio for thickness of HA coating layer shows that to obtain the highest coating thickness, the voltage value must be 80 V, the concentration 2 g/L, and the time 5 min, and this is what is available in the experiment 9. So, experiment 9 is considered as the optimal condition for obtaining the highest thickness of HA coating.

3.3. The Roughness of the HA Layer

The roughness of samples was measured only for the covered samples with the lesser amount of porosity. So, the selected samples were from experiments 6, 8, and 9. The results of Nano-roughness which was measured by AFM are shown in Table 8 and Figure 4. It is found that the highest Nano-roughness was obtained with sample 9 at 80 V, concentration 3 g/L, and time 5 min and it was 9.5263 nm. Increasing the voltage, deposit time, and powder concentration contributes to the increase in the roughness of the deposition surface. This result agrees with the results of Mohd, et al. [26].

Table 8 The roughness of HA coating prepared according to the Taguchi approach

Experiments, No.	Voltage, V	Concentration, g/L	Time, min	Nano-roughness, (nm)
6	60	3	2	6.286
8	80	2	2	1.327
9	80	3	5	9.5263

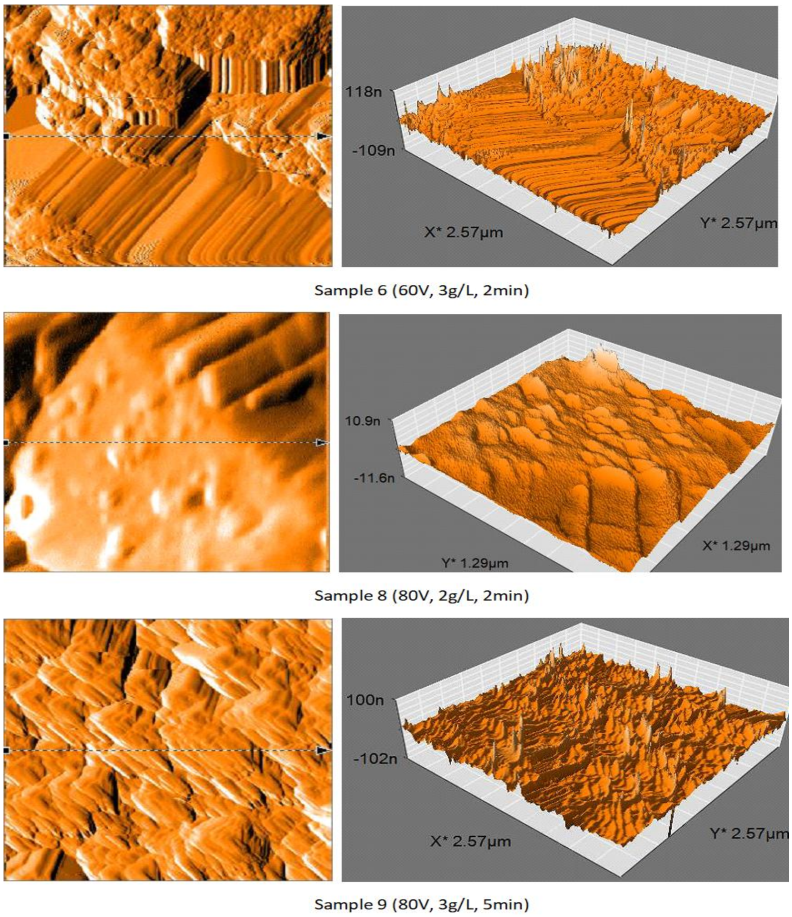

Figure 4. Two and three-dimensional features of (AFM) images for (6, 8, and 9) HA coating samples.

The roughness of the surface generates many mechanisms of the area such as mechanical interlocking, electrochemical properties, the durability of bond, and interfacial adhesion force. All these give activated mechanical interlocking last even when the chemical bonds are reduced, [27] as a result of many effects like hydrolysis, [28]. Many researchers have studied the effect of the surface roughness effect on the cells' responses to the coated implants. The results revealed enhanced proliferation of cells with increasing the roughness of samples' surfaces, [29] and this led to linked and fixation of implants with human bone, [30,31].

4. CONCLUSION

In this study, electrophoretic deposition (EPD) process was used for depositing HA Nano-powder on Ti-6Al-4V samples. The Taguchi method was utilized to select the optimum parameters of HA deposition. Morphology analysis shown that increasing the concentration of HA particles lead to decrease the amount of porosity in the coatings from 21.52 % to 0 %. Also, the porosity reduces notably with increasing the value of voltage. Full covering coating with no porosity in HA coating prepared with conditions (80 V, 3g/L, and 5min). The highest thickness of the HA coating was (18.304) prepared with conditions (80 V, 3g/L, and 5min). Depending on the Taguchi method, the (S/Ns ratio) confirms the highest thickness of the HA layer at experiment 9 with a value of 25.2509. The rank of the controlled factor on the thickness of the HA coating layer cleared that the concentration factor has the highest effect on the experiment while the voltage has the second level and the time has the lowest effect. The roughness of samples was measured only for the covered samples with the less amount of porosity. It is found that the highest Nano-roughness was obtained with sample 9 at 80 V, concentration 3 g/L, and time 5 min and it was 9.5263 nm.

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