

A review: Effects of electrophoretic deposition parameters on hydroxyapatite reinforced composite coatings

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ABSTRACT

KEYWORDS

Electrophoretic Deposition, Hydroxyapatite Coating, Metallic Substrates.

To increase the bone bioactivity of the metallic implants, ceramic oxide reinforced coatings are often deposited on implant surfaces. Various ceramic oxides such as hydroxyapatite, bioactive glass, titanium oxide, aluminium oxide, iron oxide and zirconium oxide are used for producing a real bond with the surrounding bone tissues. Among these bioactive materials, hydroxyapatite (HAp) has proved to be a promising candidate of highly reactive material. It helps to increase the bioactivity of the implant surface and possesses similar chemical, structural and biological properties to that of human bone. It will reduce metallic ion release and promoting bone-bonding ability. This review encompasses the effects of electrophoretic deposition (EPD) parameters including voltage, deposition time, dispersion medium, particles concentration, post EPD treatments and gap between electrodes on the performance of HAp reinforced composite coatings. The parameters are discussed based on the up-to-date comprehensive overview of the current research progress in the field of EPD coated HAp composite coatings for biomedical applications.

1. Introduction

Metals and their alloys are most commonly employed as orthopaedic implants because of their robust mechanical properties [1]. Titanium and its alloys, stainless steel, cobalt-chromium and magnesium are commonly used metallic implant materials [2]. They are widely used in the repair, replacement or damaged parts of the musculoskeletal system such as bones, teeth, hip replacements and implant drug delivery systems [3]. However, metallic materials are subjected to corrosion with in the body fluid environment. The surrounding tissues of the body are severely affected by the release of metallic ions [4].

To improve osseointegration with bone tissues, much research has been investigated to modify the implant surface chemically or structurally for enhancement of the bioactivity [5]. In view of these, biomaterials are commonly coated

with hydroxyapatite ($\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})$). It is a bioceramic which resembles the mineral constitutes of human bone and teeth [6]. The HAp coating acts as a barrier and decreases the release of metallic ions. Several methods for deposition on metallic implants have been investigated such as pulsed laser deposition, electrophoretic deposition (EPD) and plasma spraying [7-9]. Among these techniques, plasma

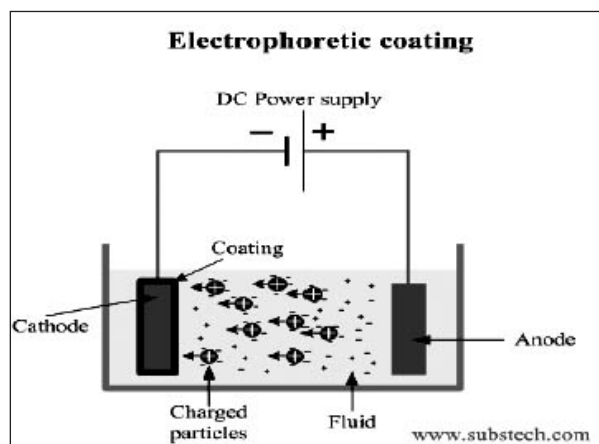


Fig. 1. Mechanism of EPD [10].

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spraying coating is widely used to fabricate bioactive coatings on metallic implants [11]. However, morphological change occurs at high temperature and results in the decline of materials bioactivity.

To solve this inconvenience, an effective technique electrophoretic deposition (EPD) has been applied for the fabrication of uniform coating. It can be performed with simple apparatus

and a low-cost method [12]. This method is adaptable for various materials and different shapes of substrates. Moreover, the coating time in EPD procedure is relatively short [13]. In particular, the EPD process could coat heterogeneous structures and complex shapes by providing uniform layers on the substrates. By altering the voltage and the deposition time, the thickness of the deposited layer can be adjusted. All these advantages make the EPD technique

Table 1

Sr. No.	Deposited Film	Reinforcement	Substrate	Voltage (V)	Time (Min.)	Dispersion Medium	Gap Between Electrodes (mm)	Ref.
1.	HAp	Bioactive glass-Chitosan	Pure Titanium	10-30	10	Ethanol	15	[15]
2.	HAp	---	Ti-6Al-4V	20	30	Ethanol	10	[16]
3.	HAp	Titanium oxide	Ti-13Nb-13Zr	30	5	Ethanol	10	[17]
4.	HAp	----	NiTi alloy	60	2	n-butanol	10	[18]
5.	HAp	Manganese	Pure Titanium	30	20	Ethanol	10	[19]
6.	HAp	Graphene oxide	Pure Titanium	30	1-5	Ethanol	20	[20]
7.	HAp	---	Pure Titanium	20	2	Ethanol	10	[21]
8.	HAp	Polyethylene Glycol	316L Stainless	30	10	Water	10	[22]
9.	HAp	Bioactive glass-Chitosan	Pure Titanium	20	10	Ethanol	15	[23]
10.	HAp	Bioactive glass	Pure Titanium	30	2	Iso propanol	10	[24]
11.	HAp	---	Ti-6Al-4V	10	10	Ethanol	20	[25]
12.	HAp	Magnesium	Ti-6Al-4V	10	5	Ethanol	10	[26]
13.	HAp	Bioactive glass-Chitosan	Pure Titanium	30	10	Ethanol	15	[27]
14.	HAp	---	Mg-3Zn alloy	20	10	Ethanol	10	[28]
15.	HAp	---	WE43Mg alloy	30	10	n-butanol	20	[29]
16.	HAp	Mg Phosphate-Zn Phosphate	AZ31 Mg alloy	20	30	Ethanol	10	[30]
17.	HAp	Chitosan	Ti-13Nb-13Zr	10	10	Ethanol	10	[31]
18.	HAp	Silk Fibron	Pure Titanium	30	10	Ethanol	10	[32]
19.	HAp	Chitosan	Ti-6Al-4V	15	5	Ethanol	5	[33]

more attractive among the researchers to modify the surface of advanced ceramic materials [14]. In this work, we review primarily EPD coated HAp reinforced coatings. Special attention is given to discuss the effect of various EPD parameters such as voltage, deposition time, post EPD treatments and the gap between electrodes on composite coatings. Based on the literature review the optimized EPD parameters are also reported.

2. EPD on Metallic Substrates With Hydroxyapatite Reinforced Composite Coatings

Table 1 shows the chronological summary of recent studies (2009-2019) for HAp reinforced composite coatings fabricated by EPD. From the analysis of the literature review range of various EPD parameters and their effects on composite coatings are identified and discussed.

3. Effects of EPD Parameters On Hydroxyapatite Reinforced Composite Coatings

The EPD process works on the principle of electrophoresis mechanism in which the movement of charged particles starts between the electrodes due to the effect of the applied electric field. The EPD method involves two types of group parameters: (a) suspension parameters (such as particle size, suspension conductivity, viscosity and zeta potential) and (b) process parameters (such as voltage, time and gap between electrodes). The effect of these parameters on HAp reinforced composite coatings are discussed in detail in the following section.

4. Suspension Parameters

Numerous parameters should be considered to understand the suspension properties such as powder surface properties, additive concentration (mainly dispersant), the influence of the type of additives and physio-chemical nature of both liquid medium and suspended particles.

4.1 Particle size

To mention the relevant particle sizes for EPD, there is no general thumb rule. However, good deposition for a variety of composite coatings has been reported to occur in the range of 1–20 μm [34]. It is worth while to specify here that

for homogeneous and smooth deposition, the particles remain stable and properly dispersed in the suspension medium [21].

4.2 Suspension conductivity

In EPD experiments, the conductivity of suspension plays a vital role. In the earlier study, it has been demonstrated that the motion of particles was low if the conductivity of suspension was higher [35]. On the other hand, the stability of particles was lost if the suspension was too resistive because of the electronically charged particles. The conductivity increases by enhancing the applied current in the suspension [23,35]. It is necessary that the conductivity lies within the suitable range for the successful assessment of the EPD process.

4.3 Suspension viscosity

To examine the dispersion rate, the viscosity property cannot be considered due to the negligible use of solid particles for the EPD process [36]. However low conductivity, high dielectric constant and low viscosity are some of the desired properties for suspension stability.

4.4 Zeta potential

In the EPD process, the zeta potential plays a pivotal role in acquiring a uniform surface charge of suspended particles. The different role of zeta potential in the fabrication of coating is as follow : (a) determining the density of the deposited material, (b) determining the movement and direction of particles in suspension and (c) resolving the interaction between particles by stabilization of the suspension [37].

4.5 Suspension stability

Electrophoresis is the occurrence of particles motion in a colloidal solution under the influence of an electric field. There are generally two types of colloidal particles in suspension. Firstly, the particles having size $1\mu\text{m}$ or less in diameter. These particles sustain in suspension for a longer period due to Brownian effect only [38]. Secondly, hydrodynamic agitation requires for particles having size more than $1\mu\text{m}$. The two major factors which characterize the suspension stability are settling rate and tendency to undergo flocculation [39]. Mori et al., [40] reported that the suspension was not stable until it shows no tendency to flocculate, make adhering deposit

and settle down slowly at the bottom of the container.

5. Process Parameters

5.1 Effects of applied voltage

In the EPD process, it is generally considered that the increase in applied voltage enhances the amount of deposition. However, higher voltage leads to film roughness, wrinkling, porosity, bubble formation and heterogeneity in the surface of the coating [41,42]. Fig. 2 depicts the variation of deposited weight with respect to voltage for different reinforced coatings. It was observed that the weight deposition was increased by enhancing the applied voltage but it can also affect the quality of the coating. Basu et al., [43] reported that the film deposited was more uniform, at a potential between the range of 25 V/cm to 60 V/cm, whereas the crack occurs in the coating if the relatively applied voltage was more than 60 V/cm. The non-uniformity of film originates at high voltage due to the anisotropy of the electric field to the substrate. Due to this reason, the deposition of particles takes place at the edges of electrodes and generates aggregates in suspension [44, 45].

It should also be noted that too low voltage does not improve the film quality. On the other hand with an increase in applied voltage leads to the formation of the porous structure. Tabesh et al., [46] analyzed that the porosity increased for HAP reinforced composite coating by increasing the voltage from 5 to 40 V/cm. Therefore, it concludes that the applied voltage should be in optimized range for the deposition of composite coatings.

5.2 Effect of deposition time

The microstructure of electrophoretically deposited HAP composite coatings is also affected by another important parameter i.e. deposition time [47-49]. The variation of time for different composite coatings are shown in Table 1. Karimi et al. [50] reported that the surface roughness of HAP films enhanced with longer deposition time due to the agglomeration of particles in suspension. Similarly, in the earlier studies [51, 52] it was also observed that the non-uniform thick layer and porosity occurs on the surface of the substrate with longer deposition time. Fig. 3 revealed that in a constant voltage EPD, with an increase in the deposition time, the effect of electric field on electrophoresis decreases, due to the generation

of particles insulating layer on the surface of electrodes. But during the starting phase of the process, the relationship between mass deposition and time was linear due to the availability of low applied field.

5.3 Effects of post-EPD treatment

For post EPD treatment of HAP reinforced coatings, different results have been reported by various researchers. The earlier studies on HAP composite coatings have shown that the heat treatment process leads to surface smoothing, compaction and reduce the interlayer spacing between the coating and substrate by removing the trapped water molecules [41, 47]. Secondly, the formation of crevices, flaky surfaces and protruding composite coating edges have

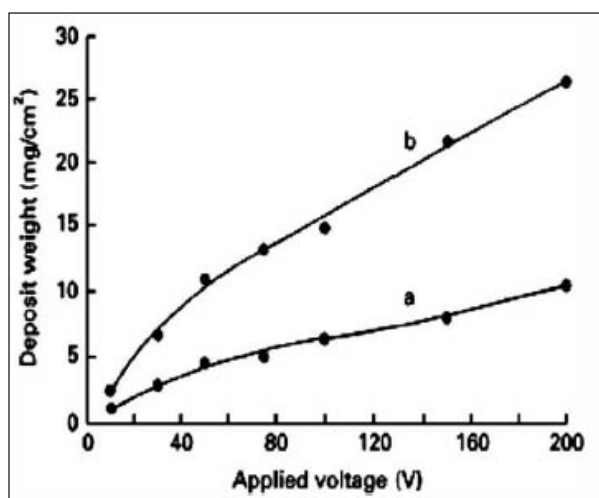


Fig. 2. Applied voltage versus the weight of deposited coating for different periods: (a) 30 s and (b) 120 s.

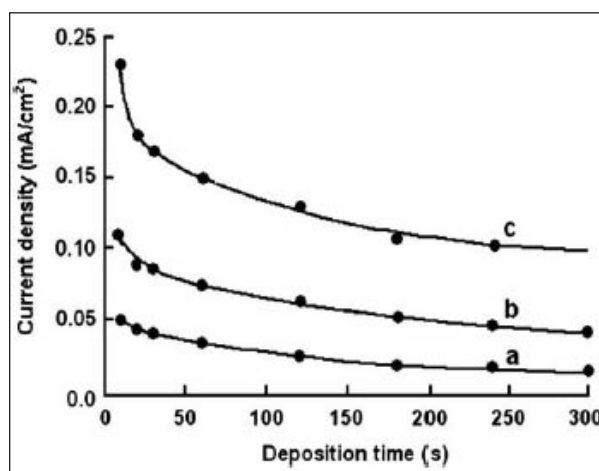


Fig. 3. Deposition of coating at different voltages (a) 10 V, (b) 20 V, (c) 30 V between current density and deposition time.

also the outcome of post EPD treatment [53]. However, at high temperature (above 1200°C), the dimension of substrate changed and results in shrinkage of coating [54]. Therefore cracks occur in the coating. During the cooling process, additional cracking occurs due to the difference in the sintered coating thermal expansion coefficients and the substrates. Other problems occur at high temperature are oxidation and phase transformations between the coatings and substrate [55]. These problems can be eliminated by selecting a suitable range of temperatures (550°C-800°C) for post EPD treatments [56]. Another alternative to avoid these troubles by the development of inorganic - polymer composite coatings. The use of polymer involves processing at low temperature and avoid the inconvenience of heat treatment process [57]. Based on Table 1 observations many of the researchers were experimented with EPD by considering the combination of ceramic-polymer composite coatings.

5.4 Effects of gap between electrodes

The gap between electrodes plays an important role in the electrophoretic deposition process. If the gap is too large there were no depositions due to the resistance offered by an electrolyte. On the other hand, if the gap was too small, there were inconveniences to flow a current flow with in electrolyte. Based on Table no. 1, many of the researcher's maintain a gap with in the range of 10-20 mm between the main and counter electrode [58]. The use of counter electrode in the EPD process provides potential to the working electrode with in the suspension medium. To keep the counter electrode dissolving in suspension medium it is usually made of inert material such as graphite on noble metal [59].

6. Conclusion

This review paper elucidated the EPD of HAP reinforced coatings on different substrates. The up-most important parameters such as voltage, deposition time, the gap between electrodes and post EPD heat treatment plays an important role during the experimentation. A significant number of research showed that the performance of HAP coatings might be improved by controlling the EPD parameters. The amount of deposit increases with increase in applied potential and leads to wrinkling, film roughness and cracks in the coating. Longer deposition times causes insufficient attachment to the substrate and

deposition of non-uniform thick coatings. At high temperature, there were chances of cracking due to the difference in the sintered coating thermal expansion coefficients and the substrates. There was no deposition if the gap between electrodes very large. From the analysis of the literature review, the most homogeneous and continuous coatings were deposited at potential difference 20 - 30 V and deposited time of 10 minutes. The temperature range lies in 550°C-800°C for post EPD treatments. The suitable gap between electrodes should be 10 mm. Based on these findings, it might be concluded that a proper combination of all discussed parameters can be effectively chosen for HAP reinforced coatings to attain high quality, smooth and homogeneous coatings. Further more, new experiments are to be conducted in which inspite of varied deposition time and voltage, the effect of several other deposition parameters like particle size, the dispersion medium, substrate, anode material, and temperature can be studied.

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