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 ($Ca_{10}(PO_4)_6(OH)$). 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

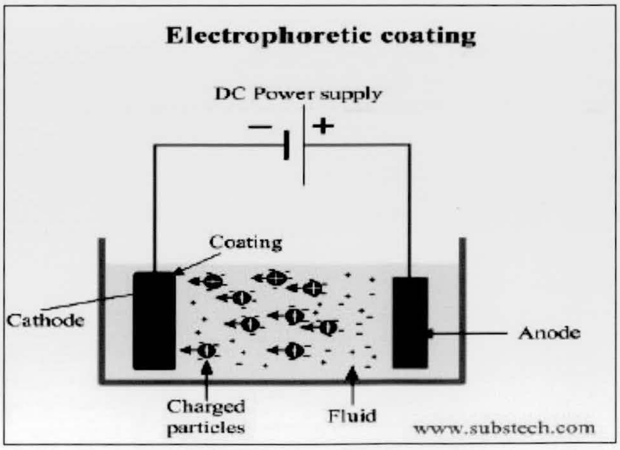

Fig. 1. Mechanism of EPD [10].

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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 with in 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 1µm 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 µm. 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 then 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

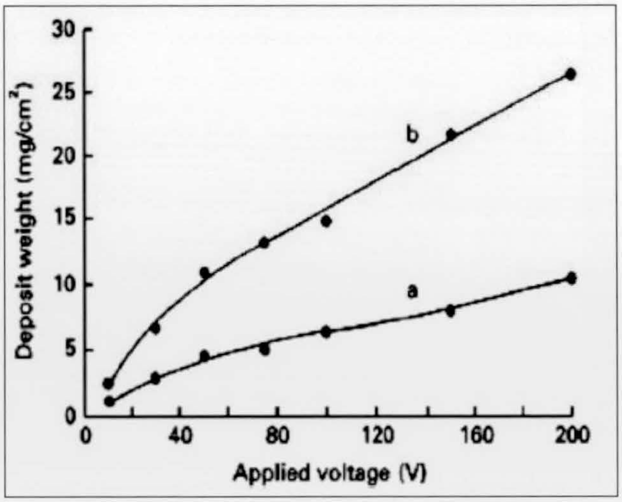

Fig. 2. Applied voltage versus the weight of deposited coating for different periods: (a) 30 s and (b) 120 s.

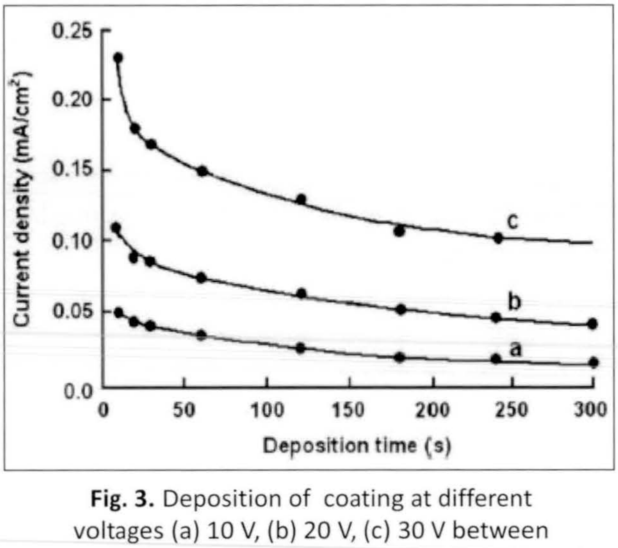

Fig. 3. Deposition of coating at different voltages (a) 10 V, (b) 20 V, (c) 30 V between current density and deposition time.

also the outcome of post EPD treatment [53]. However, at high temperature (above 1200^{0}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^{0}C-800^{0}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^{0}C-800^{0}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.

References

1. Balla, V. K., Bodhak, S., Bose, S., & Bandyopadhyay, A. (2010). Porous tantalum structures for bone implants: Fabrication, mechanical and in vitro biological properties. *Acta Biomaterialia*, *6*(8), 3349–3359. https://doi.org/10.1016/j.actbio.2010.01.046
2. Ryan, G., Pandit, A., & Apatsidis, D. P. (2006, May). Fabrication methods of porous metals for use in orthopaedic applications. *Biomaterials*. https://doi.org/10.1016/j.biomaterials.2005.12.002
3. Manivasagam, G., Kamachi Mudali, U., Rajb, B., & Asokamani, R. (2003). Corrosion and Microstructural Aspects of Titanium and its Alloys as Orthopaedic Devices. *Corrosion Reviews*, *21*(2–3), 125–160. https://doi.org/10.1515/CORRREV.2003.21.2-3.125
4. Sridhar, T. M., Eliaz, N., Kamachi Mudali, U., & Raj, B. (2002). Electrophoretic Deposition of Hydroxyapatite Coatings and Corrosion Aspects of Metallic Implants. *Corrosion Reviews*, *20*(4), 255–294. https://doi.org/10.1515/CORRREV.2002.20.4-5.255
5. Stoch, A., Brozek, A., Kmita, G., Stoch, J., Jastrz, W., & Rakowska, A. (2001). Electrophoretic coating of hydroxyapatite on titanium implants. In *Journal of Molecular Structure* (Vol. 596, pp. 191–200). https://doi.org/10.1016/S0022-2860(01)00716-5.

6. Hench, L. L. (1991). Bioceramics: From Concept to Clinic. *Journal of the American Ceramic Society*, *74*(7), 1487–1510. https://doi.org/10.1111/j.1151-2916.1991.tb07132.x

7. Liu, X., Tao, S., & Ding, C. (2002). Bioactivity of plasma sprayed dicalcium silicate coatings. *Biomaterials*, *23*(3), 963–968. https://doi.org/10.1016/S0142-9612(01)00210-1

8. Singh, I., Kaya, Cengiz., Shaffer, M., Thomas, B., & Boccaccini, Aldo. (2006). Bioactive Ceramic Coatings Containing Carbon Nanotubos on Metallic Substrates by Electrophoretic Deposition. *Journal of Materials Science*, 41. 8144-8151. 10.1007/s10853-006-0170-0.

9. Wu, C., Ramaswamy, Y., Gale, D., Yang, W., Xiao, K., Zhang, L., Yin, Y., & Zreiqat, H. (2008). Novel sphene coatings on Ti-6Al-4V for orthopedic implants using sol-gel method. *Acta biomaterialia*, *4*(3), 569–576. https://doi.org/10.1016/j.actbio.2007.11.005

10. Zhang, W., Chen, X., Liao, X., Huang, Z., Dan, X., & Yin, G. (2011). Electrophoretic deposition of porous CaO-MgO-SiO2 glass-ceramic coatings with B2O3 as additive on Ti-6Al-4V alloy. *Journal of materials science. Materials in medicine*, *22*(10), 2261–2271. https://doi.org/10.1007/s10856-011-4418-0

11. Arias, L.E.C. (2015). *Electrophoretic deposition of organic/inorganic composite coatings on metallicsubstrates for bone replacement applications: mechanisms and development of new bioactive materials based onpolysaccharides*[Ph.D. Thesis].p.1-260.

12. Boccaccini, A. R., Keim, S., Ma, R., Li, Y., & Zhitomirsky, I. (2010). Electrophoretic deposition of biomaterials. *Journal of the Royal Society, Interface, 7 Suppl 5* (Suppl 5), S581–S613. https://doi.org/10.1098/rsif.2010.0156.focus

13. ZhitomirskyI.(2002).Cathodicelectrodeposition of ceramic and organoceramic materials. Fundamental aspects. *Advances in colloid and interface science, 97(1-3), 279–317. https://doi.org/10.1016/s0001-8686(01)00068-9*

14. Zheng, X., Huang, M., & Ding, C. (2000). Bond strength of plasma-sprayed hydroxyapatite/Ti composite coatings. *Biomaterials*, *21*(8), 841–849. https://doi.org/10.1016/s0142-9612(99)00255-0

15. Zhitomirsky, D., Roether, J. A., Boccaccini, A. R., & Zhitomirsky, I. (2009). Electrophoretic deposition of bioactive glass/polymer composite coatings with and without HA nanoparticle inclusions for biomedical applications. *Journal of Materials Processing Technology*, *209*(4), 1853–1860. https://doi.org/10.1016/j.jmatprotec.2008.04.034.

16. Kwok, C. T., Wong, P. K., Cheng, F. T., & Man, H. C. (2009). Characterization and corrosion behavior of hydroxyapatite coatings on Ti6Al4V fabricated by electrophoretic deposition. *Applied Surface Science*, *255* (13–14), 6736–6744. https://doi.org/10.1016/j.apsusc.2009.02.086

17. Mohan, L., Durgalakshmi, D., Geetha, M., Sankara Narayanan, T. S. N., & Asokamani, R. (2012). Electrophoretic deposition of nanocomposite (HAp + TiO 2) on titanium alloy for biomedical applications. *Ceramics International*, *38*(4), 3435–3443. https://doi.org/10.1016/j.ceramint.2011.12.056

18. Maleki-Ghaleh, H., Khalili, V., Khalil-Allafi, J., & Javidi, M. (2012). Hydroxyapatite coating on NiTi shape memory alloy by electrophoretic deposition process. *Surface and Coatings Technology*, *208*, 57–63. https://doi.org/10.1016/j.surfcoat.2012.08.001

19. Huang, Y., Ding, Q., Han, S., Yan, Y., & Pang, X. (2013). Characterisation, corrosion resistance and in vitro bioactivity of manganese-doped hydroxyapatite films electrodeposited on titanium. *Journal of materials science. Materials in medicine*, *24*(8), 1853–1864. https://doi.org/10.1007/s10856-013-4955-9

20. Li, M., Liu, Q., Jia, Z., Xu, X., Cheng, Y., Zheng, Y., ... Wei, S. (2014). Graphene oxide/hydroxyapatite composite coatings fabricated by electrophoretic nanotechnology for biological applications. *Carbon*, *67*, 185–197. https://doi.org/10.1016/j.carbon.2013.09.080.

21. Tahmasbi Rad, A., Solati-Hashjin, M., Osman, N. A. A., & Faghihi, S. (2014). Improved bio-physical performance of hydroxyapatite coatings obtained by electrophoretic deposition at dynamic voltage. *Ceramics International*, *40*(8 PART B), 12681–12691.https://doi.org/10.1016/j ceramint.2014.04.116

22. Mehboob, H., Awais, M., Khalid, H., Siddiqi, S.A., & Rehman, I. (2014). Polymer-assisted deposition of hydroxyapatite coatings using the electrophoretic technique. *Biomedical Engineering: Applications, Basis and Communications*. *26*(6),1450073.

23. Molaei, A., Yari, M., & Afshar, M. R. (2015).

Modification of electrophoretic deposition of chitosan–bioactive glass–hydroxyapatite nanocomposite coatings for orthopedic applications by changing voltage and deposition time. *Ceramics International*, *41*(10), 14537–14544. https://doi.org/10.1016/j.ceramint.2015.07.170

24. Farnoush, H., Muhaffel, F., & Cimenoglu, H. (2015). Fabrication and characterization of nano-HA-45S5 bioglass composite coatings on calcium-phosphate containing micro-arc oxidized CP-Ti substrates. *Applied Surface Science*, *324*, 765–774. https://doi.org/10.1016/j.apsusc.2014.11.032

25. Drevet, R., Ben Jaber, N., Fauré, J., Tara, A., Ben Cheikh Larbi, A., & Benhayoune, H. (2016). Electrophoretic deposition (EPD) of nano-hydroxyapatite coatings with improved mechanical properties on prosthetic Ti6Al4V substrates. *Surface and Coatings Technology*, *301*, 94–99. https://doi.org/10.1016/j.surfcoat.2015.12.058

26. Bakin, B., Koc Delice, T., Tiric, U., Birlik, I., & Ak Azem, F. (2016). Bioactivity and corrosion properties of magnesium-substituted CaP coatings produced via electrochemical deposition. *Surface and Coatings Technology*, *301*, 29–35. https://doi.org/10.1016/j.surfcoat.2015.12.078

27. Molaei, A., Yari, M. and Afshar, M.R. (2017). Investigation of halloysite nanotube content on electrophoretic deposition (EPD) of chitosan-bioglass-hydroxyapatite-halloysite nanotube nanocomposites films in surface engineering, *Applied Clay Science*. 135, 75-81.

28. Manoj Kumar, R., Kuntal, K. K., Singh, S., Gupta, P., Bhushan, B., Gopinath, P., & Lahiri, D. (2016). Electrophoretic deposition of hydroxyapatite coating on Mg-3Zn alloy for orthopaedic application. *Surface and Coatings Technology*, *287*, 82–92. https://doi.org/10.1016/j.surfcoat.2015.12.086

29. Sankar, M., Suwas, S., Balasubramanian, S., & Manivasagam, G. (2017). Comparison of electrochemical behavior of hydroxyapatite coated onto WE43 Mg alloy by electrophoretic and pulsed laser deposition. *Surface and Coatings Technology*, *309*, 840–848. https://doi.org/10.1016/j.surfcoat.2016.10.077

30. Huang, W., Xu, B., Yang, W., Zhang, K., Chen, Y., Yin, X., ... Pei, F. (2017). Corrosion behavior and biocompatibility of hydroxyapatite/magnesium phosphate/zinc phosphate composite coating deposited on AZ31 alloy. *Surface and Coatings Technology*, *326*, 270–280. https://doi.org/10.1016/j.surfcoat.2017.07.066

31. Jugowiec, D., Łukaszczyk, A., Cieniek, Ł., Kowalski, K., Rumian, Ł., Pietryga, K., ... Moskalewicz, T. (2017). Influence of the electrophoretic deposition route on the microstructure and properties of nano-hydroxyapatite/chitosan coatings on the Ti-13Nb-13Zr alloy. *Surface and Coatings Technology*, *324*, 64–79. https://doi.org/10.1016/j.surfcoat.2017.05.056

32. Zhong, Z., & Ma, J. (2017). Fabrication, characterization, and in vitro study of zinc substituted hydroxyapatite/silk fibroin composite coatings on titanium for biomedical applications. *Journal of Biomaterials Applications*, *32*(3), 399–409. https://doi.org/10.1177/0885328217723501

33. Tozar, A., & Karahan, İ. H. (2018). A comparative study on the effect of collagen and h-BN reinforcement of hydroxyapatite/chitosan biocomposite coatings electrophoretically deposited on Ti-6Al-4V biomedical implants. *Surface and Coatings Technology*, *340*, 167–176. https://doi.org/10.1016/j.surfcoat.2018.02.034

34. Heavens, S. N. (1990). Electrophoretic deposition as a processing route for ceramics. Noyes Publications, *Advanced Ceramic Processing and Technology*. 1, 255-283.

35. Ferrari, B. and Moreno, R. (1997). Electrophoretic deposition of aqueous alumina slips. *Journal of the European Ceramic Society* . 17(4), 549-556.

36. Ferrari, B., & Moreno, R. (1996). The conductivity of aqueous Al2O3 slips for electrophoretic deposition. *Materials Letters*, *28*(4–6), 353–355. https://doi.org/10.1016/0167-577X(96)00075-4

37. Zarbov, M., Schuster, I., & Gal-Or, L. (2004). Methodology for selection of charging agents for electrophoretic deposition of ceramic particles. *Journal of Materials Science*, 39(3), 813-817.

38. Ervina, J., Ghaleb, Z. A., Hamdan, S., & Mariatti, M. (2019). Colloidal stability of water-based carbon nanotube suspensions in electrophoretic deposition process: Effect of applied voltage and deposition time. *Composites Part A: Applied Science and Manufacturing*, *117*, 1–10. https://doi.

org/10.1016/j.compositesa.2018.11.002

39. Brown, D.R. and Salt, F.W. (1965), The mechanism of electrophoretic deposition. *Journal of Applied. Chemistry*. 15, 40-48. doi:10.1002/jctb.5010150505

40. Mori, T., Nagashima, H., Ito, Y., Era, Y., & Tsubaki, J. I. (2019). Agglomeration of fine particles in water upon application of DC electric field. *Minerals Engineering, 133*, 119–126.https://doi.org/10.1016/j.mineng.2019.01.017

41. Hasegawa, K., Kunugi, S., Tatsumisago, M. & Minami, T. (1999). Preparation of thick films by electrophoretic deposition using surface-modified silica particles derived from sol-gel method. *Journal of Sol-Gel Science & Technology.* 15(3), 243-249.

42. Zhitomirsky, I. & Gal-Or, L. (1997). Electrophoretic deposition of hydroxyapatite. *Journal of Materials Science: Materials in Medicine*. 8(4), 213-219.

43. Basu, R.N., Randall, C.A. and Mayo, M.J. (2001), Fabrication of Dense Zirconia Electrolyte Films for Tubular Solid Oxide Fuel Cells by Electrophoretic Deposition. *Journal of the American Ceramic Society*. 84, 33-40. doi:10.1111/j.1151-2916.2001.tb00604.x

44. Sansone, L., Malachovska, V., La Manna, P., Musto, P., Borriello, A., De Luca, G., & Giordano, M. (2014). Nanochemical fabrication of a graphene oxide-based nanohybrid for label-free optical sensing with fiber optics. *Sensors and Actuators, B: Chemical, 202*, 523–526. https://doi.org/10.1016/j.snb.2014.05.067

45. Lee, V., Whittaker, L., Jaye, C., Baroudi, K.M., Fischer, D.A. and Banerjee, S. (2009). Large-area chemically modified graphene films: electrophoretic deposition and characterization by soft X-ray absorption spectroscopy. *Chemistry of Material.* 21(16), 3905-3916.

46. Tabesh, E., Salimijazi, H. R., Kharaziha, M., Mahmoudi, M., & Hejazi, M. (2019). Development of an in-situ chitosancopper nanoparticle coating by electrophoretic deposition. *Surface and Coatings Technology, 364*. 239–247. https://doi.org/10.1016/j.surfcoat.2019.02.040

47. Su, Y., & Zhitomirsky, I. (2013). Electrophoretic deposition of graphene, carbon nanotubes and composite films using methyl violet dye as a dispersing agent. *Colloids and Surfaces A: Physicochemical and Engineering Aspects, 436*, 97–103. https://doi.org/10.1016/j.colsurfa.2013.06.024

48. Koh, A.T., Chen, T., Pan, L., Sun, Z. & Chua, D.H. (2013). Effective hybrid graphene/carbon nanotubes field emitters by electrophoretic deposition. *Journal of Applied Physics*.113 (17), 174909.

49. Du, J., Zhang, Y., Deng, S., Xu, N., Xiao, Z., Shi, J., Wu, Z. and Cheng, H. (2013). Correlation between topographic structures and local field emission characteristics of graphene-sheet films. *Carbon.* 61, 507-514.

50. Karimi, N., Kharaziha, M. & Raeissi, K. (2019). Electrophoretic deposition of chitosan reinforced graphene oxide-hydroxyapatite on the anodized titanium to improve biological and electrochemical characteristics. *Materials Science and Engineering: C*. 98, 140-152.

51. Liu, Y., Zhang, D., Pang, S., Liu, Y. and Shang, Y., *Size separation of graphene oxideusing preparative free-flow electrophoresis*. Journal of separation science, 2015. 38(1): p.157-163.

52. Diba, M., Garcia-Gallastegui, A., Taylor, R.N.K., Pishbin, F., Ryan, M.P., Shaffer, M.S. & Boccaccini, A.R. (2014). Quantitative evaluation of electrophoretic deposition kinetics of graphene oxide. *Carbon.* 67, 656-661.

53. He, W., Zhu, L., Chen, H., Nan, H., Li, W., Liu, H. & Wang, Y. (2013). Electrophoretic deposition of graphene oxide as a corrosion inhibitor for sintered NdFeB. *Applied Surface Science*.279, 416-423.

54. Subramanian, P., Niedziolka-Jonsson, J., Lesniewski, A., Wang, Q., Li, M., Boukherroub, R. & Szunerits, S. (2014). Preparation of reduced graphene oxide–Ni (OH) 2 composites by electrophoretic deposition: application for non-enzymatic glucose sensing. *Journal of Materials Chemistry: A*. 2(15), 5525-5533.

55. Wu, M.S., Lin, Y.P., Lin, C.H. & Lee, J.T. (2012). Formation of nano-scaled crevices and spacers in NiO-attached graphene oxide nanosheets for supercapacitors. *Journal of Materials Chemistry.* 22(6), 2442-2448.

56. Drevet, R., Fauré, J. & Benhayoune, H. (2012). Thermal treatment optimization of electrodeposited hydroxyapatite coatings on Ti6Al4V substrate. *Advanced Engineering Materials.* 14(6), 377-382.

57. Dash, M., Chiellini, F., Ottenbrite, R.M. & Chiellini,

E. (2011). Chitosan - A versatile semi-synthetic polymer in biomedical applications. *Progress in Polymer Science*. 36(8), 981-1014.

58. Sarkar, P. & Nicholson, P.S., Electrophoretic deposition (EPD): mechanisms, kinetics, and application to ceramics. *Journal of the American Ceramic Society*. 79(8), 1987-2002.
59. Aziz, A., Binti, S.A., Amirnordin, S.H., Hamimah, A., Abdullah, H.Z. & Taib, H. (2012). Short Review: Electrophoretic Deposition (EPD) on Non-Conductive Substrate. In *Advanced Materials Research*, 488, 1358-136.

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