Hydroxyapatite coating techniques for Titanium Dental Implants-an overview.

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Abstract

Titanium and its alloys are the most commonly used materials for dental implants and hydroxyapatite is the most common bioactive material coated on titanium dental implants. Hydroxyapatite is from the ceramic class of biomaterials which has chemical and structural similarities with the biological apatite which forms the major inorganic portion of bone and tooth. It is not only bioactive, but also osteoconductive and non-toxic. The most interesting property of hydroxyapatite is its ability to interact with living bone tissue, forming strong bonds with the bone. Since the introduction of dental implants by Branemark in 1981, hydroxyapatite has gained attention as a preferred bioactive coating material for titanium dental implants and is still a hot topic of discussion. So, in this article, an attempt has been made to give an overview of present techniques of hydroxyapatite coatings on titanium dental implants, including their advantages, disadvantages, and limitations.

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Introduction

Titanium and its alloys are the most commonly used materials for dental implants because of their excellent biocompatibility, superior mechanical strength, and high corrosion resistance. However, they are bio-inert and do not bond or integrate with tissues (bone). A direct structural and functional connection between bone and the surface of an implant is critical for the success of implant therapy. Therefore, to facilitate implant fixation and bone growth, bioactive agents are being applied on the surface of dental implants.

Bioceramic coatings are classified into two main categories – bioinert (such as alumina and zirconia) and bioactive (such as calcium phosphate/hydroxyapatite and bioglass) coatings [1]. The bioinert ceramic coatings have the advantage of good biocompatibility and higher mechanical properties compared to the others. However, the high brittleness, high elastic modulus, and poor interaction ability with the surrounding tissues limit the use of bioinert ceramics in this field [2]. On the other hand, calcium phosphate [3] and bioglass [4], are more extensively used to treat the Ti implant due to their abilities to enhance the adhesion between the implant and the bone by interacting with the tissues.

Since the introduction of dental implants by Branemark in 1981, efforts have been made to improve osseointegration and osteogenesis by coating the implant surface with hydroxyapatite. Hydroxyapatite comes under the ceramic class of biomaterials which has chemical and structural similarities with biological apatite present in bone and tooth. It is the major inorganic constituent in bone and teeth. Titanium implants are commonly coated with it, in fact, hydroxyapatite has become an ultimate candidate for coating titanium dental implants. It is bioactive, as well as osteoconductive, and non-toxic [5]. It interacts with the bone cells forming ionic, hydrogen, and Van der Waals bonds. Hydroxyapatite can crystallize as a fine salt depending on the Ca/P ratio, the formation temperature, the presence of water or impurities, and, depending on the preparation medium, in a humid environment at relatively low temperatures [6].

Hydroxyapatite was the material of choice in the past for coating titanium dental implants and is still a hot topic of interest. In between there was a period when its use was questioned and it was about to disappear from the market because of its vulnerability to periimplantitis, dissolution, and failed interfacial adhesion between implant and hydroxyapatite [7][8]. However, advancements in coating methods have brought back the interest in hydroxyapatite coating. It is now known that the properties of hydroxyapatite can be adapted to a wide range of applications by tailoring the shape, size, morphology, and ionic substitution through different synthesis techniques [9][10]. The clinical concern with its use regarding the bonding strength between coating and the alloy/ Ti substrate can be increased by thermal spraying technology such as the hot dip method, chemical vapour deposition method, slurry method, etc. [11]. Thermal spray coating is the most efficient and commonly applied method nowadays on metallic implants due to its uniform coating layer on metal surfaces [12].

Each Implant manufacturer has its own proprietary coating process. There are many methods reported in the literature for forming hydroxyapatite coatings on Titanium implant surfaces which can be broadly classified under two categories – Pyroprocessing methods (e.g. Plasma spraying, sol-gel method, electron beam sputtering, ion beam sputtering, etc.), and Hydroprocessing methods (e.g. Cathodic electrolysis method, thermal substrate method, etc.) [13].

Some of the commonly used hydroxyapatite coating techniques for titanium dental implants are briefly discussed in this article. Among those techniques, only plasma spraying is commercially approved by the Food and Drug Administration (FDA), USA for biomedical coatings on implants due to its excellent coating properties over other coating methods [14]. However, this technique is limited nowadays because of some limitations mentioned below under the heading 'Plasma spraying.' So, alternative coating approaches have been extensively developed and tested the sol-gel dip coating and electrochemical deposition techniques have relative ease of production, the ability to apply a uniform coating over complex geometric shapes, easy setup at room temperature, the potential to deliver exceptional mechanical properties, corrosion resistance, and adhesion strength due to their nanocrystalline structure [15].

Active ingredients Entrapment methods of nano- and micro-encapsulation to achieve the desired controlled release of the ingredients are being actively researched [16][17]. Conventionally it was done by binding them to the substrate or coatings or mixing them into the coating material and applying the mixture to the implant surface. However, recently interest has shifted to a new method of entrapment of the active ingredient within another substance by microencapsulation or nanoencapsulation. Coating with a bioactive growth factor (Hydroxyapatite-Bone Morphogenetic Protein-2 coating) has been determined to enhance the relationship between an implant and the bone to which it is attached [18][19]. BMP-2 (Bone Morphogenetic Protein-2) enhances the osteogenic activity of osteocytes, accelerates osteoblast differentiation, and further promotes bone formation. It can also induce dentin and post-implant healing. The incorporation of bacteriostatic and bactericidal agents in the hydroxyapatite coating is widely being performed [20][21][22][23][24][25]. Another hot topic that has received increased biomedical research attention in recent years is the influence of pore size and porosity of HA coatings [26] on 3D-printed [27] Ti scaffolds which have inherent porous structures. Besides the implementation of coatings, titania nanotube formation using anodization has gained interest nowadays[28][29][30].

Thus, the literature is full of studies done on hydroxyapatite coatings on titanium dental implants using various methods and techniques. However, for a quick review of all the existing techniques for hydroxyapatite coatings, there are only a few studies done. Therefore, the aim of this article is to give a quick overview of existing techniques for hydroxyapatite coatings on titanium dental implants.

Sol-Gel Dip-coating Technique

The process starts with the dispersion of the precursor particles (fine nano-sized particles) in an aqueous or alcohol solution forming a colloidal suspension (the sol). Then the addition of catalysts to the sol promotes polymerization reactions involving hydrolysis and polycondensation forming a gel [31][32].

The dip-coating method (DCM) consists of a well-controlled immersion process. The five sequential steps are: (1) immersion of the pretreated titanium implant in the coating mixture at a specified rate; (2) keeping the implant immersed in the mixture for a specific amount of time; (3) lifting the implant out of the mixture or lowering the mixture container, which results in the formation of a wet liquid film of the coating on the surface of the implant; (4) draining off the excess liquid for the coating application from the implant surface; and (5) drying the coating layer by evaporation of the solvent under ambient conditions, thus forming a thin film on the substrate which can eventually be hardened by conventional or laser sintering [33][34][35].

Advantages of this technique are [36][37][38][39][40][41]; i) It allows the deposition of nanocomposite hydroxyapatite films reinforced with nanotubes or of doped films containing elements such as F, Sr, Ag to provide various actions, ii) Though, the speed and time of immersion are determinant factors of film thickness, generally it allows for the attainment of very thin films, and thus maintains the original surface roughness of implants. Thick or irregular coating layers are not desirable because they might modify the surface roughness and could negatively interfere with cell adherence, differentiation, and consequently affecting osseointegration.

However, there are some limitations [42][43][44][45][46][47] of DCM like, i) The high-temperature sintering cycle has been judged to be unsatisfactory because of its adverse effect on the mechanical properties of the titanium-like grain growth and loss of the wrought structure of both the commercially pure Ti and Ti-6Al-4V substrates may occur, ii) Concern about the bond strength between the hydroxyapatite coating and the substrate which is not high enough to ensure that interfacial failure will not occur during the lifetime of the implant, iii) Its use on an industrial scale imposes a complete technological change, and iv) It requires accurate control of the stability of the sols and of all the stages of coating formation.

Electrochemical Deposition (ECD)

According to C.A.D Rodriguez, Electrochemical deposition is a process by which a thin and tightly adherent desired coating of metal, oxide, or salt can be deposited onto the surface of a conductor substrate by simple electrolysis of a solution containing the desired metal ion or its chemical complex [48].

It is the conventional well-known process based on the principle of electrolysis, that uses electrical current to reduce the cations of a desired material from an electrolyte and coat those materials as a thin film onto a conductive substrate surface. In other words, it is a well-known method to produce in situ thin, metallic, inorganic, or organic coating by the action of an electric current on a conductive material immersed in a solution containing a salt of the metal to be deposited. Other terms like electrodeposition and electroplating are also used for this method.

Advantages of ECD [49][50][51][52][53][54] are: i) Performed at room temperature, is a rapid, straightforward, and versatile method; ii) Good control of the coating material's thickness, uniformity, crystallinity, and stoichiometry; iii) Thus, capable of delivering an HA coating on titanium implants with satisfactory homogeneity, thickness, and bonding strength; iv) No phase transition problems as found with PS-fabricated HA coatings; v) The morphology of the HA coating can be modified by adjusting the parameters like the pH, temperature, deposition voltage and immersion time during the coating process;

vi) Anodic oxidation of Ti can improve the tear strength between HA coatings; and vii) Ti substrates and HA coatings can be doped with Mg to change the properties and facilitate the control of the dissolution rate of the HA coatings.

Limitation of Cathodic ECD: i) Poor adhesion of the HA layer to the implant's surface[55].

Electro-phoretic Deposition: (EPD)

In this method, electrodes are immersed in a colloidal suspension, and a potential is applied between the two electrodes which results in the movement of the charged suspended particles of HA from the solution to the oppositely charged electrode (substrate), followed by the deposition of these particles on the electrode and the formation of a uniform coating up to 2.00 mm thick [56][57][58].

Advantages of EPD are [59]: i) Low-cost (operates in mild aqueous/organic environment at low temperatures and pressures, with excellent efficiency, and short processing time) ii) Non-line-of-sight coating process, iii) Very likely to produce coatings of uniform dimensions, iv) versatility of the process (film thicknesses can vary from nano- to millimetre-scale).

Limitations are [60][61]: i) Poor Adhesion of coatings because of challenges in morphology control, and rates of deposition that are influenced by the time dependency of the parameters that include colloidal concentration, composition, electric field effects, pH, electrophoretic mobility, etc.); and ii) Lack of understanding of the interaction of complex factors that influence EPD.

Biomimetic coating

Biomimetic means it mimics nature or biology and near physiological conditions are employed for deposition of the apatites [62][63]. This is a relatively new method in which heterogeneous nucleation and crystal growth of the coating occurs, which has bonelike properties [64]. For this, pretreatment of the implant surface is done by using either alkaline (NaOH) or acidic (HF or HCl) solutions or heat treatment and then, the substrate is immersed in simulated body fluid (SBF) at body temperature (37 C) and physiological pH (7.4) [65][66] It is important that the SBF contains an ion concentration similar to human blood plasma[67]. The obtained results after several weeks in SBF are CaP-based coatings deposited on the Implant surface.

The advantages of this method are [68][69][70][71]; i) it allows for the production of homogeneous HA coatings on porous implants, and ii) incorporation of functional and biological agents, such as growth factors, in HA coatings is possible because of the near physiological conditions employed. However, there are some limitations of this method, such as: i) difficulty in controlling the thickness, the formation rate, and the quality of the coating, and ii) unacceptable bonding strength of the coating thus developed. [72][73].

Plasma spraying (PS)

This is basically a process of spraying molten or heat-softened material onto a surface. A type of thermal spraying that has been established as the most commonly used commercial coating technique for the fabrication of CaP/ HAP coatings on Ti–6Al–4V implant surface because the process is time convenient, highly repeatable, efficient, and simple[74]. In this method, the generation of a direct current arc in the plasma torch, which consists of a cone-shaped cathode and a cylindrical anode is done [75][76]. Then, HA material in the form of powder is fed into the spray gun, heated to a semi-molten or molten state (the plasma flame temperature being in the range of 6,000 -16000 degree centigrade), and then propelled to the implant surface with a high impact velocity (up to 400 m/s) which results in the coating material particles get flattened on the implant surface in the form of splats/lamellae with a large surface area and a thickness in the micrometre range [77][78].

Based on the differences in the pressure conditions, there are variants of plasma spraying techniques like atmospheric plasma spraying (APS) employed under atmospheric conditions, low-pressure plasma spraying (LPPS), also known as vacuum plasma spraying (VPS), operates at 3-7 kPa, and very low-pressure plasma spraying (VLPPS) operates at 50-200 Pa [79][80][81].

Although PS is the preferred method in most cases, there are some limitations like i) Excessively high temperature and the subsequent rapid cooling procedure generate a large amount of amorphous HA, ii) Variations in the structure of HA occurs, iii) Line-of-sight nature of the spraying process due to which the interior of the 3D porous structure is not accessible for coating and, iv) It Limits (but does not rule out) the possibility of drug release applications [82][83].

Moreover, the current trend in research is to manufacture thin (≤ 50 μm) HA layers, which do not alter the surface texture of titanium implants and contain less residual stress, reducing the risks of delamination. Such low thicknesses are barely attainable by the Conventional plasma-spraying technique [84].

High-Velocity Suspension Flame Spraying Technique (HVSFS)

It is a new method of acquiring the nanostructured dense surface coatings on the titanium implant surface by spraying the nanoparticles with hypersonic speed for which the powder is dispersed in aqueous or organic solvent and fed axially into the combustion chamber of a modified High-Velocity Oxyfuel spray-torch[85][86]. This has been developed because the conventional High-Velocity Oxygen Fuel (HVOF) spraying processes are not suitable to achieve nano-particle coatings. The powder is processed in the form of a suspension (in aqueous or organic solvent) to solve the problems related to the handling of powders composed of nanosized particles, and by doing this its feeding is also facilitated with quite simple thermal spray techniques[87][88].

Examples are the suspension plasma spraying (SPS) or the solution precursor plasma spraying (SPPS) methods. The liquid solvent used in this technique permits to inject particles in the thermal flow which is then heated, accelerated, and sprayed onto the substrate. Thus, as compared to conventional plasma spraying, SPS and SPPS are more complex methods because fragmentation and vaporization of the liquid solvent control the coating buildup mechanisms [89][90][91].

Physical vapour deposition (PVD)

The vacuum deposition methods where materials are evaporated or sputtered, transferred in the form of atoms, molecules, or ions, and deposited onto the substrate surface in the form of thin films are known as PVD techniques[92][93].

PVD technology includes a wide range of techniques, like cathodic arc deposition, electron beam deposition, evaporative deposition, pulsed laser deposition, ion plating, ion beam deposition, magnetron sputtering, etc [94][95][96]. For the deposition of bioactive hydroxyapatite-based coatings on titanium implants, sputtering techniques are the ones found to be more convenient than other PVD techniques.

Sputtering involves a process of ejecting neutral atoms from a target surface using energetic particles (argon ions) which can be easily accelerated towards the cathode by means of an applied electric potential, hence bombarding the target, then transferred and condensed on the surface of the substrate to form a coating [97]. There are several variants of the sputtering method the simplest model being the diode plasma, which consists of an anode and a cathode between which appropriate potential difference is applied which ionizes argon gas and creates a plasma discharge, the argon ions then get attracted and accelerated toward the sputtering target and displace some of the target atoms which results in electron emission that subsequently collide with gas atoms to form more ions that sustain the discharge [98].

Magnetron sputtering is the most common approach to the sputter deposition technique. It is a promising option to overcome the problems such as delamination and low bond strength that may arise with plasma spray methods[99]. Magnetron sputtering allows operation at lower voltages and pressures because it uses magnets to form a magnetic field parallel to the target which allows trapping of the secondary electrons near the target[99][100]. This induces more collisions with neutral gases and increases plasma ionization.

Advantages of the Sputtering technique are[100][101]: i) High deposition rate, ii) High purity, iii) Excellent adhesion of the coating to the substrate, iv) Ability to produce dense and thin coatings, as well as provide good bond strength and, v) Ability to coat different surface geometry.

Limitations of the Sputtering technique[101]: i) Several factors can affect the properties of the coating like the target and substrate condition, process parameters, and the equipment used, ii) Several fundamental factors about the process are still poorly understood.

Pulsed laser deposition (PLD):

It is a physical vapor deposition (PVD) technique where a high-power pulsed laser beam is focused inside a vacuum chamber to strike a target of the material that is to be deposited[102].

In this, a laser having a high-power density and narrow frequency bandwidth is used as a source for vaporizing the desired material and there is almost no restriction on the target material to be used. This technique can be considered when other techniques have failed to make the deposition and have been used to synthesize the nanotubes, and nanopowders[103][104][105].

This technique has been shown to produce various bioceramic coatings including stoichiometric HA onto metallic substrates under controlled experimental conditions. These HA-coated metal implants are expected to be superior in function [106][107].

Chemical vapour deposition(CVD)

CVD is similar to PVD in the basic process of utilizing vapour and generating a thin film on a substrate. It differs from PVD in that it does not generate vapour from a solid or liquid source in a vacuum chamber. Instead, vapours or gasses are introduced into the chamber from an external source which then gets deposited on the substrate surface in multiple directions in the form of non-volatile solids through a chemical reaction[108][109]. Thus, unlike PVD, it involves a chemical reaction of vapour-phase precursors and the chemical reactions of precursors occur both in the gas phase and on the substrate which results in better adhesion to the surface. Another significant difference from that of PVD is that it is not limited to line-of-site application and thus, can be theoretically applied to any area the coating gas can get into, coating gas will coat all areas of a part including threads, blind holes, and interior surfaces [110]. Activation energy is often required to initiate the reaction and the gaseous by-products produced during the reaction are periodically pumped out[111].

Conclusion

The well-established technique of coating hydroxyapatite layer on titanium dental implants is Plasma Spraying Technique. In recent years, with the introduction of nanotechnology and 3D-printing of implants, interest has shifted towards manufacturing thinner HA layers and the techniques which allow for the production of homogeneous HA coatings on porous implants. This is not possible with the Conventional plasma spraying technique because of which many newer techniques have been proposed, and even the plasma spraying technique is being modified. Another factor that has compelled us to think about alternative techniques is the need for the incorporation and controlled release of bacteriostatic, bactericidal agents, and other active ingredients in the coatings. The main limiting factor in the use of newer techniques like the Sol-Gel Dip-coating technique and electrodeposition technique is a complete technological change as compared to the thermal spray processes.

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