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Review of Various Hydroxyapatite Coating Methods on SS316L Foam for Biomedical Application

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Abstract: Metallic biomaterials such as 316L stainless steel (SS316L) are widely used as an implant to replace the function of damaged bone, especially in hip or knee applications. However, many of them fail during a short period or have complications. The biocompatibility issues are the main factor that caused this failure. Thus, coating the SS316L with bioactive and biocompatible material is one of the promising techniques to enhance the biocompatibility and lifetime of the implant. This paper provides an overview of the SS316L foam coated hydroxyapatite (HA). Various methods of HA coating such as sol-gel, dip coating, electrophoretic deposition, plasma spraying, and pulse laser deposition applied on SS316L foam and their coating characteristics were investigated based on recent literature. SS316L foam coated HA using different coating methods were compiled and their basic properties were reported. Therefore, this paper will benefit future works on SS316L foam coated HA in a biomedical application.

Keywords: Stainless steel, SS316L foam, hydroxyapatite, coating, biomedical

1. Introduction

A biomaterial is an essential material used as an implant material in biomedical applications. The biomaterial can be defined as any substance, natural or man-made, comprising part of a living structure or biomedical system that performs, improves, or replaces a natural function of the human part [1]. The 316L stainless steel (SS316L) foam is one of the metallic biomaterials used and adapted for a medical application, especially for replacing the damaged hip or knee bone tissue due to its excellent mechanical properties and relatively inexpensive compared to other metallic biomaterials. It is an inviolable substance designed to interact with biological systems that can operate successfully after implantation. SS316L foam needs to own empty or fluid-filled voids (porous) with a solid metal matrix structure. Metal foam can be classified into two types; open-cell and closed-cell. Interconnected open voids (channel) are called open-cell. Meanwhile, voids that are separated by solid walls are known as closed-cell [2]. The pores size of foams for biomedical application should have interconnected macro-porosity with pore size >100 mm or micro-porosity with pore size < 20 mm. Multi-scale porous foams involving both micro and macro porosities can perform better than foams with only one-dimensional porosity [3].

Despite possess excellent mechanical properties as biomaterials, the SS316L reported capable of localised corrosion after a long period of in vivo implantation that disturbed the proliferation of human body fluid [4]. On top of that, the

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bioinert properties of SS316L facilitate no bioactive properties. Therefore, an appropriate combination of chemical, mechanical, physical, and biological properties is needed to design well-established biomaterials that can prolong the implant lifetime and avoid loss of implant function. In order to address these issues, one of the promising approaches is the development of bioceramic coating on SS316L. This approach provides a remedy for corrosion issues and promotes biocompatibility and bioactivity properties to SS316L, which is crucial as an implant material [5].

Hydroxyapatite (HA) with the chemical formula $[Ca_{10}(PO_4)_6(OH)_2]$ is a bioceramic that is widely used as a coating material due to its superior biocompatibility and promote bone in-growth once in contact with living tissue [5]. It is a good replacement material for bone and dental implantation [6] since the chemical and biological properties of HA are similar to the minerals contained in human bones and teeth [5,7]. HA coating can provide excellent biocompatibility and bioactivity properties to metallic implants [8]. HA is a bioceramic that is mostly used in orthopaedic and dental applications. Thus, HA various coating methods mainly to enhance the biocompatibility and bioactivity properties of SS316L foam have been investigated. There are many types of coating methods available such as sol-gel coating, dip coating, plasma spraying, electrophoretic deposition (EPD), and pulsed laser deposition (PLD) [9]. Therefore, the present article deals with a review of HA-coated SS316L foam methods to offer wide-ranging available information on this material based on recent works of literature.

2. Coating Method Preparation

There are various types of coating methods that can be used for surface modification of SS316L foams. The hydroxyapatite (HA) coating method review could provide comprehensive information on methods preparation and their advantages and disadvantages.

2.1 Sol-Gel Dip-Coating

Chemical and physical homogeneity can be improved through the sol-gel method by blending on a molecular scale among multiple coating deposition procedures [10,11]. Due to small particle sizes with large surface areas and can create uniform fine grain structures, these methods also reduce the heating temperatures [12,13]. Additional advantages of this method include the use of several chemical routes (aqueous and alkoxide) and their ease of application to complex shapes using various coating techniques such as spray coating, dip, and spin. The sol-gel method combined with the dip-coating technique has been widely used in the coating of biomaterials to improve adhesion [14]. Dip coating is a three-stage method: dipping, removal, and drying, as shown in Fig. 1. The substrate is dipped into the coating solution and then withdrawn at a constant speed, allowing for precise thickness control and no waste. By using a lower coating temperature, the sol gel-dip coating method can produce a thin adhesive layer of coating without serious cracking [15]. The capacity to regulate the purity and chemistry of the coatings is one of the special advantages of sol-gel dip-coating, followed by calcination [16]. This deposition method has the advantage of producing a wide range of coatings with a high degree of consistency [17]. The substrate is usually removed vertically from the required coating solution in this method, resulting in a complicated process involving gravitational drainage, simultaneous drying, and condensation responses. Temperature, moisture, and airflow are just as important as the pH solution and removed velocity. They all have an impact on the film's parameters [9, 18, 19]. Evaporation of solvents (primarily ethanol and water) forms thin films, which concentrate non-volatile species in the system and cause aggregation and gelation. The resulting film is determined by the size, structure, withdrawal speed, substrate surface, and pH of the precursors.

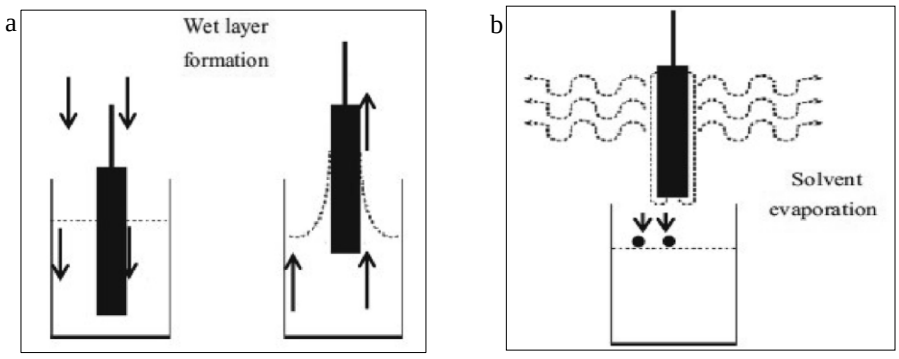

Fig. 1 - Sol-gel dip-coating technique; (a) Immersion and withdrawal; (b) Drying [20]

Apart from being an excellent synthesis method, the sol-gel technique has several drawbacks. The main drawbacks are gel drying and extreme volume shrinkage during gelation, removal of unwanted residuals (hydroxyls and organics), and the presence of large pores that have limited its use in the industry [21]. Only 0.5 μm is the maximum coating thickness for crack-free coating. Besides, trapped organics during the heat cycle would cause coating failure. Heat expansion mismatch is also a problem in recent advances in highly delicate sol-gel substrates. However, there are still

spaces for enhancement in the technique, and further research should be carried out to enhance this extremely prospective biomaterial coating method [22]. Due to the compatibility of biomaterials and coating methods, sol-gel may be the best HA coating method for metal substrates. For example, the coating process can be done at room temperature to prevent HA decomposition at high temperatures. Therefore, a few analyses for the characterisation of coatings have been investigated from previous researchers to improve the quality of sol-gel deposited HA layers. Table 1 below shows a research study related to the sol-gel dip-coating process of HA on SS316L.

Table 1 - Research study related to the sol-gel dip-coating process of HA on SS316L

Precursor for Sols and Other Materials	Operating Conditions	Outcome	Year	Ref.
• Calcium nitrate • Phosphorus pentoxide • Ethanol • SS316L	• Constant dipping rate of 7 mm/s. • Sintering at 150 and 500°C. • Coated for 5 times.	• HA coated samples show better corrosion resistance and better implant properties as compared to uncoated SS316L.	2019	[23]
• Calcium acetate monohydrate • Phosphoric acid • Calcium acetate monohydrate • 1,2 - ethanedio ethylenediaminetetra • Acetic acid • Triethanolamine • Polyvinyl alcohol • Distilled water • SS316L	• Dip-coating immersing (85 mm/min) and withdrawal (40 mm/min) rates. • Spin coated at 2000 rpm for 60s in air. • coated 5, 15 and 30 times. • Annealed at 850°C for 5h in an oven.	• Both dip coating and sol-gel methods are suitable for the fabrication of HA films on the SS316L substrate.	2016	[24]
• HA • Sago starch • Acetone • Aquades • Water • SS316L	• Aquades use 16, 18, 20g. • Dip-coat immerse for 20s. • Sintering temperature 600, 700 and 800°C for 1h.	• Thickness of HA coating composition increased in the range of 42 - 144 µm when aquades use increase. • Maximum shear strength obtained was 11.78 MPa for temperature 600°C using 16g aquades.	2016	[25]
• HA • Sago starch • Aquades • Water • SS316L	• HA use 8, 10, 12 g • Dip-coat immerse for 2, 6, 10s. • Sintering temperature 800°C for 1h.	• Increased HA addition and dipping time will increasing the thickness of the coating HA. • Maximum shear strength obtained in this research is 0.24433 MPa for 10s dipping time, HA used 12g.	2016	[26]
• Calcium nitrate tetrahydrate • Phosphorus pentoxide • Ethanol • SS316L	• Dipping speed of 10 mm/min. • coated 3, 4 and 5 times. • Sintering temperature 800°C for 1h.	• The coating developed by 4 times dip have minimum surface roughness, adhesion strength and maximum micro hardness. • The coating developed by exposing substrate for 4 times dip in sol offered a better corrosion protection.	2015	[27]

The study of the sol-gel deposition of HA coatings from Dominguez-Trujillo et al. [28] proved an excellent fitting between HA coatings and Titanium (Ti) substrates owing to mechanical interlocking and potential chemical bonding. The bioactivity of the metal surface and the avoidance of metal ion discharge (more corrosion resistance) are both indicated by the HA coatings. The layer also has a homogeneous morphology, with no cracks and/or discontinuities. The

sol-gel method has recently attracted much attention due to its well-known intrinsic benefits in producing glass, glass-ceramic, and ceramic powders. These include the ability to generate size particles, low processing temperature, homogeneous molecular blending, enormous flexibility to create nanocrystalline powders, and thin films [14,29]. In addition, the sol-gel method is a simple way to cover a surface and allows for the preparation of high-quality HA thin films on metal substrates [30-33].

Sol-gel coatings are a method of applying a protective coating layer to the surface of metallic biomaterials used in biomedical applications [14, 34, 35]. Even though the sol-gel coating process has been widely used recently due to its excellent coating properties, it should be improved in some way to achieve the best results. Important aspects of the sol-gel coating process, such as adhesion and delamination, should be given more attention [36,37]. Furthermore, sol formation parameters such as solvents, ageing, pH, and temperature must be studied in order to improve the process and coating properties [38, 39]. In addition to traditional coating methods such as spinning, dip coating, and spraying, new research and optimisation for sol-gel routes are required to provide reliable coating layers. These sol-gel techniques could help improve the quality of SS316L foam coated HA for biomedical applications.

2.2 Electrophoretic Deposition (EPD)

EPD is a method of developing thin or thick films and coatings by coating particles in a suspension onto an electrode under the influence of an electric field. [40-43]. At electrode, deposition of charged particles occurs by a coagulation process. Controlling the deposition parameters and the size and form of the ceramic powder allows for coating thicknesses ranging from 1 mm to more than 100 μm thick [40, 44]. The parameter can affect the EPD process are deposition time, electric field, pH, particles concentration, and the conductivity of the suspension [45-47]. EPD is a flexible, low cost, non-line of sight coating method for depositing uniform thin films on surfaces with complicated shapes or surface morphologies [48]. The general principle of EPD is a two-step process in which colloidal charged particles or molecules in suspension migrate towards an oppositely charged electrode under the influence of an externally applied electric field (electrophoresis), then particle or molecule accumulation and coagulation to form a compact deposit at the electrode surface (deposition) [49]. The EPD setup consists of a power supply coupled to two parallel electrodes, a working electrode and a counter electrode, immersed in a particle solution in its most basic form. As shown in Fig. 2, EPD contain four steps; i) dispersion, ii) electrochemical charging, iii) electrophoresis, iv) deposition. In principle, any particulate solid (ceramics, polymers, metals, and their composites) or biological entity (proteins, cells, enzymes, etc.) can be used in EPD as long as the particles can be stably suspended and produce a substantial surface charge in interaction with the solvent. Under the influence of the applied electric field, the charge helps to prevent sedimentation caused to agglomeration and to promote high electrophoretic mobility.

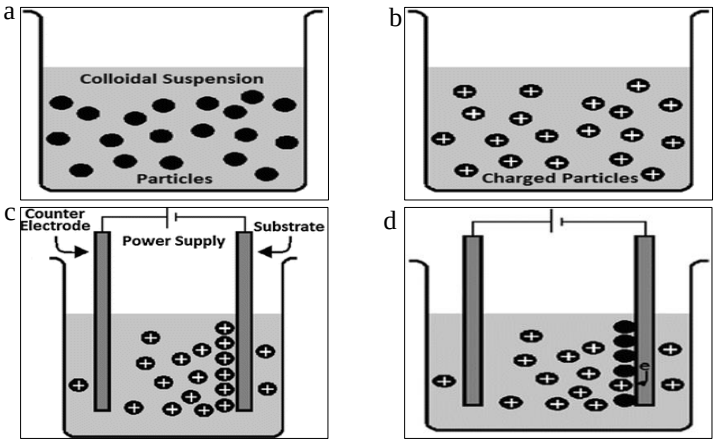

Fig. 2 - Four steps of EPD; (a) dispersion; (b) electrochemical charging; (c) electrophoresis; (d) deposition [50]

EPD advantages of being a simple process that only requires essential equipment, having a short deposition period at room temperature with controllable thickness, being relatively inexpensive, having good adhesion strength of deposits, and having the ability to co-deposit various particles [51]. The movement of charged particles in a suspension is the basis for this method. Charged particles begin to move in the produced electric field and deposit on electrodes when voltage is applied to a substrate (anode and cathode). The process is usually done at room temperature, and it can also be used to make uniform deposits on large product sizes with high microstructural homogeneity for a variety of equipment and complex shapes [52,53]. The morphology and thickness of the coatings can be tailored and controlled by adjusting the applied voltage and deposition time. It is necessary to use a stable colloidal solution containing charged particles that move when an electric field is applied to deposit homogeneous coatings [54]. This method can also be used to apply

various coatings to metallic substrates, such as ceramics, polymers, and composites [55,56]. However, the primary challenges of EPD are the need to develop stable suspensions and the fact that coatings cannot be applied directly until the deposit has been sintered for densification [51,57]. EPD requires conductive substrates, which limits the types of substrates that can be used. Despite being a wet process, EPD allows for easy control of the thickness and surface morphology of the deposited layer by adjusting the deposition time and applied potential.

On the other hand, water cannot be used as the liquid medium in the EPD method because the applied voltage in water causes the generation of hydrogen and oxygen gases at the electrodes, which has negative effects on the quality of the deposited films poses a safety risk. In contrast to other colloidal processes, this is considered an inherent limitation of EPD [58]. This method is one of the most promising methods to produce HA coatings bonding to the substrate [59]. As a result, it is expected that by combining the mechanical properties of SS 316L with the bioactivity of HA, a better orthopaedic implant will be produced. Table 2 shows a research study relate to HA deposited on SS316L metal.

Table 2 - Research study relate to HA deposited on SS316L substrate

Electrolyte and Other Materials	Operating Conditions	Outcome	Year	Ref.
• Capiz shells • Ethanol • Acetone • SS316L	• SS316L stainless steel substrates were used as anode and cathode. • The solution was stirred for an hour at 60°C. • Voltage variation of 25V and 50V 15 min. • Withdrawn from the solution with the speed variations of 0.1, 0.5, and 1 mm/s. • Sintering temperatures were 750, 850, and 950°C. • Coating time 10, 20, and 30 min.	• More homogeneous and free of cracking at a voltage of 50V and the withdrawal speed of 0.1 mm/s. • Layer density increase when the sintering temperature and voltage used was increased. • The best results were obtained by applying a 50V, a withdrawal speed of 0.1 mm/s, and a sintering temperature of 850°C.	2020	[60]
• HA • Aquades • Ethanol • SS316L • Carbon	• Carbon as anode and SS316L as a cathode. • The voltage used is 40, 50, and 60V. • 30 min deposition time and coating time.	• The thickness of HA layer on substrate increased as the time deposition increased. • Thickness HA coatings can decrease the corrosion of SS316L. • 50V and deposition time 30 min was suitable for a bone plate.	2019	[7]
• Calcium nitrate tetrahydrate • Diammonium hydrogen phosphate • Ammonia • Ethanol • SS316L • SS314L	• SS316L acts as cathode and SS314L as an anode. • Stirring was maintained at 700 rpm and left undisturbed for 2 h. • Coatings were carried out at a constant voltage of 40V for 5-10 min at room temperature. • The distance between the electrodes was 1 - 2 cm. • It is sintered in a vacuum furnace at 800˚C for 2 h.	• Developed HA coating on SS316L would be ideal for orthopaedic applications. • Results show increasing in roughness and compressive stress. • This method offered better corrosion protection than uncoated SS316L.	2016	[61]

Continue Table 2

• Cuttlefish lamella bone • Ammonium dihydrogen phosphate • Aquades • Methanol • Ethanol • Triethanolamine • Platinum • SS316L	• The voltage used was 30, 40, and 50V. • Platinum as anode and SS316L as cathode. • Sintered at temperature 250˚C for 2h and followed by 800˚C for 2h.	• HA layer on the surface of SS316L with the best optimum voltage at 40V samples and has the best corrosion rate.	2016	[62]
• Dihydrogen phosphate • Clamshell • Ammonia • Ethanol • Nitric acid • Carbon • SS316L	• Carbon as anode and SS316L as a cathode. • Substrate calcined at 800˚C for 2h. • The concentration of HA 0.025 and 0.075 M • pH 3, 4, and 10.	• The thickness of HA deposits increases with the concentration of HA. • The thickness of HA deposits decreases when pH increase. • Corrosion rate result shown that HA coating can reduce the corrosion rate of SS316L substrate.	2015	[63]

The study of electrophoretic deposition from other researcher has added other material with a combination of HA to coat on SS316L like bio-mimetic minerals [64], chitosan and titania [65] silver-strontium [66], gelatin [67], and chitosan [68]. All this research does not sinter their substrate after EPD process as sintering can lead to cracking of coating and all this research have a gap between anode and cathode substrate 1 cm as if the gap is bigger will cause increasing of resistance that can cause less deposited material on cathode [68-70]. Though the apatite coating degrades over time due to exposure to the harsh environment, toxic metal ions leach out of the underlying SS316L bio-implants. Therefore, surface treatment of implant with bio-mimetic minerals and bio-ceramic coating are applied to survive through long-term implant situations [64]. If there is a presence of water in the suspensions, the weight of the coatings increased linearly with a time of deposition, increasing the electrical conductivity of the deposited film resulting in higher solid concentration that causing cracks and rough surface, hence lower corrosion resistance [64]. In another study, the incorporation of strontium improves the bioactivity of HA. The incorporation of silver in the HA provides an antibacterial effect against a broad spectrum of bacteria [68, 71, 72]. Gelatin (protein-based polymer) as a gelling agent or binder and it has been shown that its thermogelation supports the adherence of deposit to a substrate during the EPD process [68].

In order to improve deposition process control, many attempts have been made to describe the underlying mechanisms of EPD. While the EPD stage is well understood, the exact mechanism of particle aggregation at the electrode surface is still under debate; currently, the double-layer distortion model is most accepted. The deposition yield can be correlated with various deposition parameters, regardless of the detailed mechanism. Hamaker's law can be used to develop useful mathematical expressions for EPD kinetics [73]. The thickness of which is determined by the EPD parameters, and the morphology of which depends on the deposited substrates [74]. A stable suspension of charged particles is a prerequisite for successful EPD. The metal-organic framework particle must be able to produce a stable colloidal suspension, and organic solvents must be used to avoid any side reactions as it is a crucial requirement in EPD technique [75]. According to the deposition mechanism, it is evident that deposited coating has weak adhesion and cohesion that need post-sintering [56]. However, oxidation of substrate and thermal stresses in the coating and thermal decomposition of HA are the consequences of sintering that lead to cracking [76]. Problems caused by sintering could be overcome by adding polymers like gelatin and chitosan to HA coatings or other suitable additional material [68, 67, 77]. Because of that, it is necessary to research optimising the parameters of EPD (voltage and coating times) to know the optimum parameters [78].

2.3 Plasma Spraying

Plasma spraying processes were developed in the 1980s as a practical method used in bioceramics coating on metal substrates, particularly for hip prostheses with HA coating, thus eventually becoming the industry's choice to utilised it [79-81]. The broad utilisation of calcium phosphate bioceramics as bioactive coating materials may be because it has rather similar composition with mineral components of bone, thus promoting speedy implant fixation [82-84]. Still, the study mentioned that hydroxyapatite it bulk form has a negative impact as it contributes to low fracture toughness but not in disperse powder form as utilised as coating material in the plasma spraying process [85]. In the present, plasma

spray is still widely known as a promising coating technique capable of implementing new coating properties as demand to biomedical metallic implants [86,87]. Generally, this process is applied towards emerging demands of orthopaedic implants for corrosion-resistant, wear-resistant and high temperature resistant thermal barrier coatings [88,89]. As the pioneer, plasma spray was first introduced in the production of calcium phosphates coatings such as HA and β-TCP owing to the fact that it is simple to implement with low production cost [22,90]. Moreover, the capabilities of the plasma spray technique in bonding between HA coatings to bone directly alongside metallic substrates were so favourable as showing positive and promising results in bone recovery applications [91]. The idea of the coating should have low porosity, strong, cohesive strength, good adhesion to the substrate, a high degree of crystallinity, and high chemical and phase stability besides having to obtain bioactivity and durability [92].

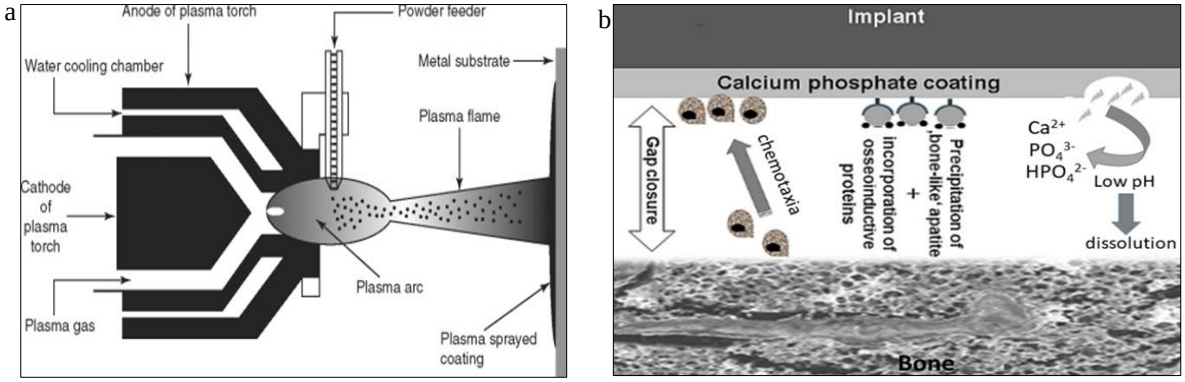

Fig. 3 - (a) Plasma spray schematic diagram [95]; (b) schematics of the interaction between implant and living bone cells, intervened by a thin calcium phosphate coating [101]

This technique was carried out by applying the melted materials solution and spray onto the substrate surface using high temperature and pressure on an electric arc to melt and showers the dried HA powder to the metallic substrates [93]. As schematically illustrated in Fig. 3 (a), the dry powder of feedstock with fully crystalline HA and particle size ranging from 10 to 90 μm in diameter is transformed into molten by applied heat using a thermal plasma jet [94]. Plasma torch comprises several important components to accomplish the coating process of metallic implants, which are cone-shaped thoriated cathode made up of tungsten, cylindrical anode made up of copper and the powder feeder. The plasma forming gas or arc gases such as argon, helium, hydrogen and nitrogen flow through the annular space between two electrodes and the arc is then initiated by high-frequency discharge before loops out of the nozzle of the torch as a plasma flare [95]. Then, after being sufficiently heated and accelerated by plasma jet, the melted materials are sprayed onto the targeted substrate surface. This coating provides a protective layer that can prevent corrosion, wear, or high temperatures [96,97]. Once impact, the droplets of molten particles cool down and straightaway solidify by means of heat transfer to the underlying substrate and consequently forming a coating consist of lamellae as the result of accumulation [98]. The HA coating is atmospherically plasma sprayed using various spray power and stand-off distance (SOD) [99]. The end of the method would be when the plasma spared coating is formed by the build-up of successive layers of particle droplets flattened upon impact as graphically shown in Fig. 3 (b), and thus the coatings display layered structures [100,101]. Table 3 shows some important process parameters involved in plasma spraying of HA onto the metallic implant, such as titanium, stainless steel and other metal alloys.

Table 3 - General process parameter in coating different metallic implants via plasma spray technique

Raw material	Metallic implant	Process parameters	Outcome	Year	Ref.
HA/β-TCP	Titanium	• HA/βTCP particle size 100-200μm • Plasma voltage: 62V • Feed rate: 35 g/min • SOD: 10 mm • Heat treatment at 500-1000°C	• Coatings on titanium substrate are uniformly obtained and well adherent. • β-TCP phase is almost converted to α-TCP due to high temperature and cooling rate during plasma spray processing.	2018	[102]

Continue Table 3

HA	Stainless steel	• Plasma Current: 500 amp • Ar plasma gas flow: 90 scfh • H_2 plasma gas flow: 15 scfh • Ar powder feeding gas flow: 13.5 scfh • Number of passes: 12 • Spray distance: 75 mm • Speed of spray gun: 200 mm/min • Speed of specimen table: 100 rpm	• The corrosion resistance of single-layer coated far more superior compare to double layer HA/TiO_2 coated.	2016	[103]
HA	Mg alloy	• Primary gas flow: 37.6 V/m^3h^{-1} • Secondary gas flow: 7.05 $V/m^3\ h^{-1}$ • Voltage: 500 U/V • Current: 70 I/A • Rotational speed: 35 n/r min^{-1} • Thickness: 0.1mm	• The coating result demonstrates a lower corrosion rate and better bioactivity than magnesium alloy. The coating significantly improved the hydrophilicity of the Mg alloy.	2018	[104]

In the other study conducted by [105], the SS316L substrate is prepared by curing in acid (8% HF and 40% HNO_3) for 1 min and rinse in acetone to remove the oxide layer and degreased. Then, the substrate is grit-blasted for ~30 s with pressure at 6 bar. Any residual grit on the substrate surface is removed by air-blasting and finally cleaned with alcohol. Principally, the investigation of the adhesion between the metallic substrate and HA coating is one of the crucial concerns when performing plasma spraying techniques [106]. Besides, numerous studies also recognised that the plasma-assisted deposition techniques outshine as coating technique with rapid, well-controlled, economically advantageous and highlight the developed way to coat almost any substrate with those materials that exhibit the well-defined congruent melting point [107]. Even though numerous researchers widely studies coating metallic implants by plasma spray technique, this process faced some inevitable limitations, which is the deformation of hydroxyapatite structure due to high temperature [108]. Throughout the plasma spray process, it is necessary to be in high-temperature conditions; however, it changes the compositional and structural transformation of HA and β-TCP. It transforms into amorphous calcium phosphates (ACP) phases such as tetra-calcium phosphate (TTCP), α-tricalcium phosphates (α-TCP), and β-tricalcium phosphate (β-TCP) [109]. Therefore, post-heat treatment can enhance the microstructure, leading to an increase in both cohesive strength and adhesive of coating [110].

2.4 Pulsed Laser Deposition (PLD)

Numerous surface modifications techniques of metallic implants were utilised to offers novel approaches in enhancing its clinical versatility [111-114]. A widely known method adopted for metallic orthopaedic application to promote bone regeneration on metallic implants is to create a composite coating comprising bioceramic components on their surface [115-117]. PLD is a coating technology used to coat various substrates employing highly accelerated laser beams in the vacuum environment [118-120]. The PLD method's significant benefit is that it can deposit uniform, pure, crystalline and stoichiometric hydroxyapatite films with controlled surface properties of the substrate layer [121]. The PLD method has been used to deposit high-quality films of materials for more than a decade. As shown schematically in Fig. 4 (a), (b), the working principle of PLD is straightforward which utilising pulses of laser energy to form a thin layer of coating [122,123]. Then, the vaporised coating materials containing neutrals, ions, electrons and other elements will expand rapidly away from the target surface at velocity typically ~106 cms^{-1}. Finally, a thin layer of film growth appears on a substrate surface upon which some of the plume material recondense [124]. Despite that, in real practice, the situation gets tricky with numbers of variables needed to attend to as it affects the film's properties produced, such as laser fluence, background gas, and substrate temperature [125-127]. Therefore, investigators concentrated on finding optimisation of deposition conditions to achieve deposited layers' good physical and chemical properties [34,128]. As for the coating material, the bioceramic coatings on the metallic implant can avoid toxic metal ions released from the implant surface into the surrounding environment; thus, the system's entire bioactivity and bone mineralisation were ensured better and improving [129]. Multiple studies have reported successful incorporation of calcium phosphates bioceramic (HA) or its precursor's coatings on various metallic substrates as reviewed in Table 4, namely Ti-6Al-4V, stainless steel, NiTi and other bio metallic alloys, which results in positive effects on various aspects of bone regeneration.

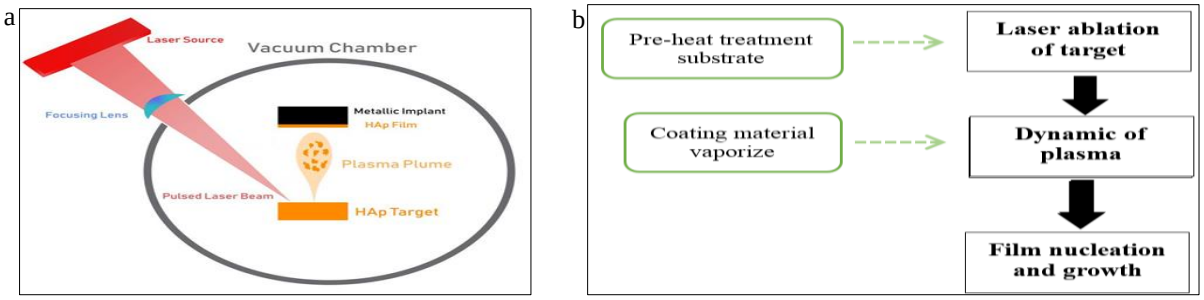

Fig. 4 - (a) Schematic diagram of typical PLD set up; (b) Block diagram of PLD process [130]

Table 4 - PLD on different types of metallic implant in biomedical applications

Raw material	Metallic implant	Process parameters/conditions	Outcomes	Year	Ref.
Hopeite (hydrated zinc phosphate)	Ti64	• Deposition time:60 min • Gas flow:30 cm^3/min • Energy fluence:2.5 J/cm^2 • Laser energy:100 mJ • Pulse frequency:10 Hz • Pulse width:5 • Substrate temperature:500°C • Target-substrate distance:40 mm	• X-ray diffractograms prove the existence of hopeite coatings on Ti64 substrate. • In vivo performance reveals rapid apatite growth on the surface of hopeite coated specimen. • Outstanding osteointegration achieved.	2020	[131]
HA	ss316L	• Repetition rate:50 Hz • Pulse duration:25 ns • Rotating angle:45° • Substrate temperature:570°C	• Uniformly coating surface obtained with increasing in laser energy from 300 to 500mJ. • Increasing in crystallinity with increasing in laser energy confirmed by AFM and XRD.	2013	[132]
HA	αAl_2O_3/ ss316L	• Wavelength:248 nm • Repetition rate:5 Hz • Pulse energy:~400 mJ, 5000 shots • Target-substrate distance:45°/5 cm • Laser fluence:1.416 J/cm^2	• HA films were grown below 400°C less stable compare to at 700°C, which is more crystalline. • Uniform PLD coatings achieved, flat morphology. • EDS reveals peaks corresponding to coating-substrate constituents.	2014	[133]
HA	Au/ Al_2O_3	• Wavelength:1064 nm • Repetition rate:10 Hz • Pulse duration:8 ns • Focal length:50 cm • Target-substrate distance:80 mm	• By controlling the coating composition, great potential biological responses were successfully achieved • This including cell adhesion and viability.	2020	[134]
HA	NiTi	• Wavelength:1064 nm • Laser:50 kHz • Pulse duration:100 ns • Laser power:40 W • Repetition rate:20 kHz • Focus length:200 mm	• Ability to performed PLD to NiTi substrate to generate osteoinductive surface • Have advantages to complex micro-nano surface structures of bone.	2018	[135]

Continue Table 4

HA	SS254	• Wavelength:532 nm • Rotating angle:45° • Substrate temperature:600°C • Target-substrate distance: 40 mm • Deposition pressure:10^{-6} mbar • Energy fluence:2.5 J/cm^2 • Pulse frequency:10 Hz • Pulse width:5 ns	• AFM and SEM micrographs confirmed excellent coverage of coating from the PLD technique in columnar with an increase in surface roughness. • Performing postdeposition annealing to achieved desired crystallinity.	2016	[136]
HA	Mg/Ca alloy	• Wavelength:532 nm • Energy fluence:1.5 J/cm^2 • Target-substrate distance:3.5 cm • Deposition pressure:10^{-5} mbar • Substrate temperature: ~400°C	• The recommended substrate temperature is at 200-300°C. HA decomposed if higher than that. • Show good corrosion resistance in SBF.	2018	[137]

3. Conclusion and Future Development

This article aimed to provide a comprehensive overview of preparing and developing biomedical implant coatings. Based on the review of various coating methods that have been done, the best coating method that produced good characterisation of HA coating on stainless steel is obtained by plasma spraying method while the worse characterisation of fabricated stainless steel is obtained by sol-gel coating. The HA coating thickness produced by plasma spraying is 50 μm which is advisable in biomedical implantation because coats less than 50 μm thick are quickly dissolved, and coats more than 50 μm may suffer delamination [138]. Moreover, the HA deposition was uniform while it covered the entire surface of stainless steel foam. Proper growth of HA crystals and almost 99% purity of HA coating formed on a stainless steel surface. Furthermore, the sol-gel coating is a less recommended coating method because the thickness of HA coating obtained is less than the advisable thickness. The HA coating layer is also deposited within cavities that act as sol solution reservoirs during dip coating and resulting in a thick-coated area that is more prone to cracking due to excessive drying and sintering strain [139].

However, it was evident that numerous parameters can affect the performance of HA coatings, such as preparing methods, preparation parameters, quantity, and quality of coating elements. It means that there is still a need to perform thorough research to optimise the parameters mentioned above to enhance the biocompatibility performance of HA coatings and decrease the number of failures or infections in orthopaedic implantation. There is still high demand to develop ionic doped HA coatings onto implant materials for future development since applying doping elements into the HA crystals can improve the biocompatibility of implants.

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