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Nanomaterials coating for bio-implant applications: a re-analysis

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Abstract. Bio-implants are inserted in the body to support the natural part of body either fractured or partially failed. Therefore, bio-implant should be compatible with the body; must not produce harmful effect to body tissues or organs. As body environment is corrosive in nature; the implant should have high corrosion resistance. Also, it should have high strength and low percentage elongation. Additionally, implant material must allow the growth of tissues so that high bonding between the implant and body tissue can be achieved. Based on all the above-mentioned requirements for being a good implant material, titanium, steel, cobalt-chromium alloys, etc. are most widely used as implants. To further enhance the mechanical and biological properties of implants, different types of coating and surface modifications are done. Coating thickness, type of coating, and coating deposition techniques significantly affects the properties of an implant. In the present work, effects of the above-mentioned parameters are studied on the mechanical and biological properties of the implants. It was observed that the biocompatibility and wettability of polymer coatings were relatively less in comparison to ceramic and composite coatings. On the other hand, ceramic coatings were highly biocompatible and wettability was also high. For metallic coating, biocompatibility was less because of high reactivity. But, metals (like Bi) that form protective oxide layer on the surface of implants exhibits very good biocompatibility and mechanical strength.
Keywords: biological properties; coating; Implant; mechanical properties

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Introduction

Bio-implants are used to support or replace the partially functional or non-functional part of a living body. Use of implants started in the ancient times itself when glass eyes, gold filled teeth, wooden leg were used to replace the failed body parts [1]. Now, with the development of technology, use of implants have been increased tremendously. Now implants for almost all the body parts such as eye, teeth, breast, hip, knee, etc. are available. Fig.1 shows the use of implants for human body [2].

Primary function of implant material is to support the fractured body parts, so the strength of implant materials should be high enough to bear the body weight [3]. As an implant is inserted inside the living body. So, along with having sufficient mechanical properties, biological properties of an implant material should also be similar or better than the natural part. A schematic diagram of hip implant and various properties required in an implant material are shown in Fig. 2 [4].

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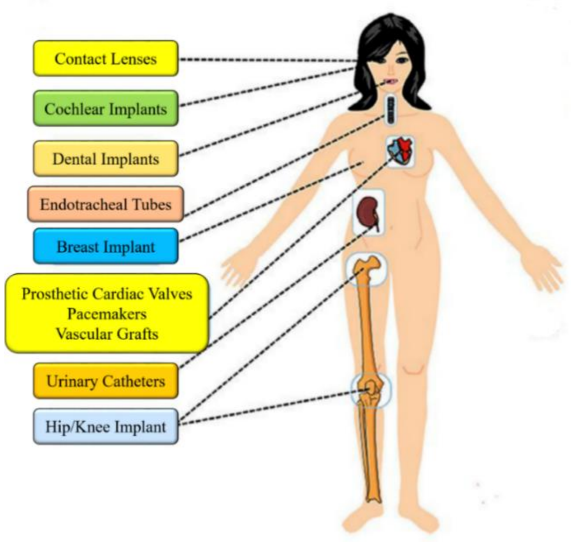


Fig.1. Use of implants

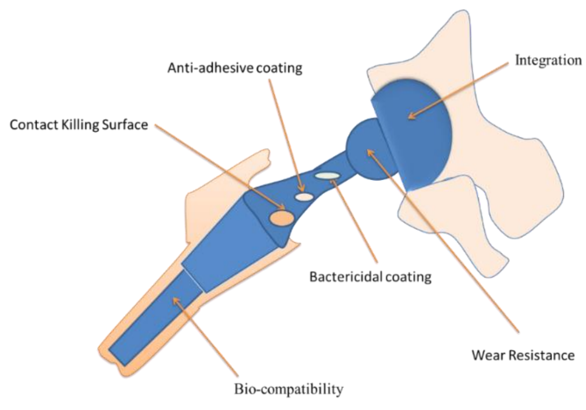


Fig. 2. Schematic diagram of hip implant and various properties required in an implant material

Considering the criticality of implant, the selection of appropriate material for implants become very important [5]. Some of the properties that are required in a good implant material are as follows:
1. It should have strength comparable to natural bone;
2. Percentage elongation should be very low;
3. It should be biocompatible;
4. It should not release toxic elements;
5. It must support the growth of tissue;
6. Ability to resist viral infections or contamination.

Most commonly used materials for implants like titanium (Ti), Ti alloys, stainless steel 316L (SS 316L), Co-Cr alloys, tantalum alloys, composites, ceramics, and polymers are given in Table 1 [6]. These materials are widely used in medical equipment because of their good corrosion resistance, and high strength [7,8].

Table 1. Implant materials and their applications

Biomaterials	Applications
Stainless Steel	Hip and knee joint implant, dental implants, spinal, heart valve, hip nail, bone plate, shoulder prosthesis.
Titanium Alloys	Cochlear implant, dental implants, orthodontic implant structure, artificial heart valves, bone and joint replacements, and pacemakers.
Cobalt Chromium Alloys	Total joint replacements (hip and knee) mini plates, bone plates, screws, dental implants, orthopaedic implants.
Alumina	Acetabula, femoral components, vertebral spacer and extensor, orthodontic anchors, dental implants, and artificial complete joint replacement.
Zirconia	Replacement of hips, knees, tendons, ligaments, periodontal disease treatments, and bone fillers.
Calcium Phosphate	Dental implants for throat repair, skin treatments, dental fillings, jawbone reconstructions, orthopaedic coatings on implants, and facial surgery.

Problems with implant materials

Infection of the implant by microbial contamination on implant surfaces or adjacent tissue is one of the main problems of implant failure, as microorganisms can readily adhere to and then grow on implant surfaces [9,10]. This type of transplant infection often occurs in different parts of the human body. In addition, the open wound created during the implant placement process creates the right conditions for microbes to adhere, grow and multiply. Although the host's immune response is activated, this response is often insufficient to clear the infection around the implantation site [11,12].

Infections associated with biomedical devices and implants (e.g., catheters, heart valves, and hip and knee implants) remain a major risk for their long-term usage despite extensive research and development efforts [13]. Bacteria can apparently cling to and colonise a wide range of biomedical devices, resulting in a difficult-to-treat infection. Infections, lengthy hospitalisation, difficult revision operations, transplant failure, patient misery, financial load, and even death are some of these. As a result, the necessity to eliminate germs or limit bacterial colonisation is growing.

Infections, corrosion, wear, excessive inflammation, severe toxicity, poor osseointegration, and foreign body effects are all factors that shorten the life of orthopaedic implants [14]. Infection may cause the patient to suffer for a long time and eventually die. Corrosion and wear wreak havoc on a structure's structural integrity. However, the implants cause toxicity. Our immune system may mistake the implant material for a pathogen. System, causing chronic inflammation and foreign body reactions, which can lead to death. A local and most effective technique for resolving these issues and extending the life of implants is to coat them with biocompatible and bioactive material. Coatings transport biologicals directly to the injury site, allowing them to work more precisely than pharmaceuticals.

Requirement of coatings

The principle of bone on-growth or in-growth is linked to the optimal shape (design) of an implant, as well as its material, surface topography, and chemistry [15]. The materials typically utilised for orthopaedic implants, stainless steel and titanium (alloys), have differences or rather benefits in strength, ductility, stiffness, or cell responsiveness, and must be considered according to the medical necessity [16,17]. The implant's design can compensate for mechanical flaws, but host cells and bacteria have different reactions to the material [18]. The disadvantages of electropolished steel (smooth surface) implants are imaging artefacts, possible implant migration, release of toxic/allergic ions such as cobalt, chromium, nickel, and higher infection risk, according to preclinical research and clinical experience, whereas the disadvantages of rougher titanium implants are removal complications due to better osteointegration [19,20]. Polymers, such as PEEKs (polyetheretheretherketone), are primarily employed in the field of spine surgery as a radiolucent alternative to metallic implants, however due to biomechanical constraints when compared to titanium or stainless steel, a general application has yet to be established [21,22]. Because solid metal implants, particularly those made of cobalt-chrome or steel, have a lower degree of osteointegration than pure titanium, zirconium, and/or titanium-based coatings, they can be withdrawn more quickly (temporary implants) [23]. The surface chemistry (hydrophilic vs. hydrophobic) and the surface topography, specifically the roughness, are the primary determinants impacting osteointegration, in addition to the material's biological compatibility. If osteointegration is to be enhanced, specific material selection, surface modification, or extra coatings are required (permanent implants) [24]. Chemical, physical, and biological requirements are the most important for this purpose [25]. For an implant coating to promote osteointegration, biocompatibility is an obvious necessity. Despite the fact that various materials and surface modifications are utilised to maximise implants, comparative studies proving the evidence for "the best" material, surface condition, or coating are still lacking. Another need is that the material's properties be preserved during use, and that the biological interaction in the body has no detrimental consequences. Coatings can address the tendency of wear and corrosion in an in vivo environment, which can result in particle loosening or the release of hazardous substances [26]. Implant-derived wear particles activate cells of the innate and adaptive immune systems, such as macrophages and lymphocytes, triggering an inflammatory response marked by the release of cytokines, chemokines, and growth factors, which can lead to osteolysis and bone loss due to increased osteoclast activity and necrosis [27]. In the creation of new coatings or the translation of authorised coatings into new applications, this in vivo interaction of the material must be taken into account. Another criterion is that the coating stay on the surface of the implant at least until it reaches its desired position and can perform its function.

Temperature resistance of the implant material to the temperature introduced during the coating or surface-structuring process is a production requirement. For example, the temperature during a plasma spray process for HA can exceed 500 °C, whereas it can exceed 1000 °C for calcium phosphates [28,29]. Laser structuring raises the surface temperature well over the boiling point of the substrate (e.g., 3260 °C for titanium). The influence of temperature, particularly for metallic implants, can cause metallurgical changes, necessitating further biomechanical considerations. Surface alteration (blasting, etching) and porous coatings can cause fracture initiation, which has been shown to affect biomechanical needs for fatigue and bending strength. Other obvious needs are the ability to sterilise the final implant without causing any unwanted effects, and the ability to meet the requisite shelf life without causing undesirable changes in the features over time.

Bioactive coatings

The demand to replace and restore damaged tissues has increased in tandem with the general population's longevity. This is especially true in dentistry and orthopaedics, where prosthetic implants have restored structure and function. These surgeries have been incredibly successful, and they have improved the lives of a lot of individuals. For the most part, this success can be credited to the body's ability to integrate with the implants. In exchange, implant materials and surface qualities that most effectively trigger bodily responses have been adopted. The implants' surface can be altered in a variety of ways, either directly or by applying a coating.

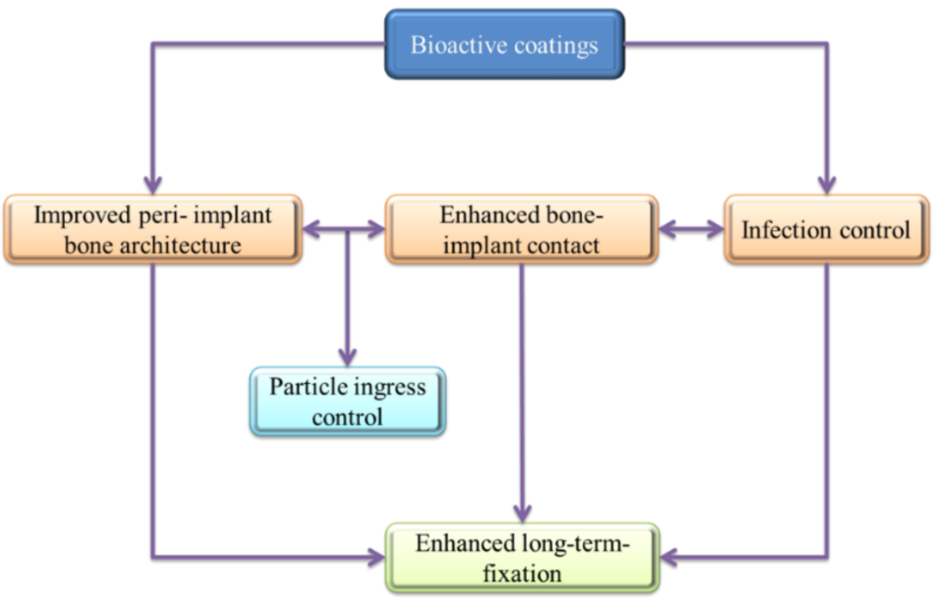


Fig. 3. Potential mechanisms of action of bioactive coatings

The goal of improving long-term mechanical fixation of the implant is based on the assumption that a stable bond between the implant and the host bone will contribute to long-term clinical success. Even if a bioactive coating does not allow an implant to perform its intended function for a longer amount of time (i.e., enhance survivorship), it would be useful to reduce the time required to achieve fixation. The implant surface can be affected by topography, chemistry, and surface energy in a broad sense. Potential mechanisms of action of bioactive coatings are shown in Fig. 3.

All implants are viewed as alien objects by the body, they stimulate a biological response in the form of a "non-self" material to counteract any negative consequences. This reaction is influenced by both implant-related and host-related factors. Implant-related aspects that are influenced by the physical, chemical, and biological features of the implant surface are discussed here. Tissue-related parameters such as the implant site, patient gender and age, tissue integrity, and systemic diseases are not discussed in any depth. The reader should be aware, however, that the biological response to a specific implant is not expected to be consistent across all patients. The features of the implant's manufacturing material, in general, impact the body's reaction to it. The word bio-inert refers to the absence of a reaction to the implant. In reality, when bio-inert materials are introduced in body, they interact very less with the surrounding tissue, resulting in a minimum response. As a result, a fibrous membrane forms around the implant has no effective bonding with body [30]. Bioactive materials, on the other hand, initiate a chain of events that leads to the production of new extracellular matrix, which, in the best-case scenario, forms in intimate contact with the implant and results in mechanical fixation.

Effect of different types of coatings

There are various types of coating materials such as polymer, metals, ceramics, composites, etc. Depending on the nature of coating materials and their interaction with the substrate, properties of a coated implant are very much different from the uncoated implant. As, polymers have high resistance to corrosion and are hydrophobic in nature. Polymer coating improve the corrosion resistant reduces the swelling. Metals are more reactive in comparison to polymers, so metallic coating reacts with oxygen to form metallic oxides. Metallic oxides are very less reactive and biocompatible in nature. Metallic coating also forms strong bond with metallic substrate. So, metallic coating improves the strength and biocompatibility of the implant. Ceramic and composite coatings also improves the biocompatibility and other properties of implants. Effect of different type of coatings on various properties of implants are studied below.

Bond strength. Bond strength is the measure of bonding between the coating material and substrate. High bond strength is desirable because high bond strength enhance the coating stability and life. The variation in bond strength of Ti alloy with different coating materials is shown in Fig. 4. It can be observed that the significant bond strength was obtained in all the coated Ti_6Al_4V implants. Ghaleh et al. investigated the effect of akermanite (tricalcium magnesium silicate) coating on the bond strength of coated Ti6Al4V titanium alloy [31]. Bond strength of 40.5 MPa was observed. Xue et al. investigated the effect of diopside ($CaMgSi_2O_6$) coating on the bond strength of coated Ti_6Al_4V titanium alloy [32]. Bond strength of 32.5 MPa was obtained. Yi et al. done the bredigite coating on the Ti_6Al_4V alloy and investigated the bond strength of the coated titanium alloy [33]. Highest bond strength of 49.8 MPa was observed in the Ti alloy coated with bredigite. Li et al. coated the titanium alloy Ti_6Al_4V with monticellite hardystonite ($Ca_2ZnSi_2O_7$) and investigated the bonding strength [34]. Bonding strength obtained was around 33.4 MPa. Liang et al. studied the effect of baghdadite ceramic coating on the bond strength of Ti_6Al_4V titanium alloy [35]. Lowest bonding strength of 28 MPa was obtained. Wu et al. investigated the effect of ceramic coating of $Sr_2MgSi_2O_7$ on the titanium alloy substrate on the bonding strength [36]. Bonding strength of 37 MPa was observed. So, it can be concluded that the ceramic bredigite forms strong bond with the titanium alloy Ti_6Al_4V and hence the strength and life of coating will be more in comparison to other ceramic coatings. As, the implants are intended to work satisfactory for 5–15 years, so the bredigite coated titanium alloy will be preferred.

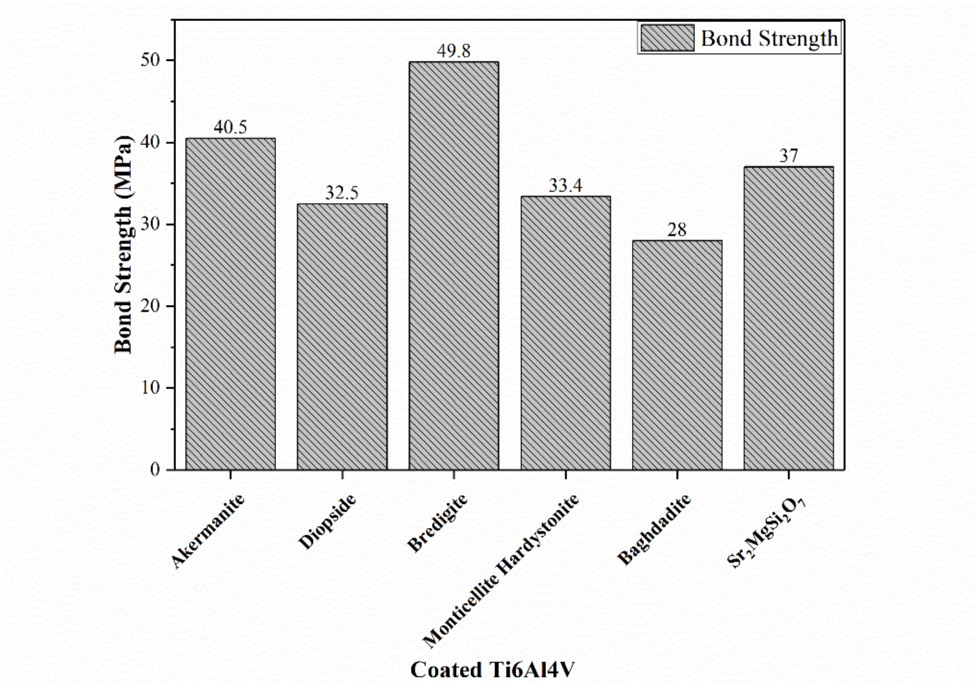


Fig. 4. Variation in bond strength of Ti alloy with different ceramic coating materials. Based on data [31–36]

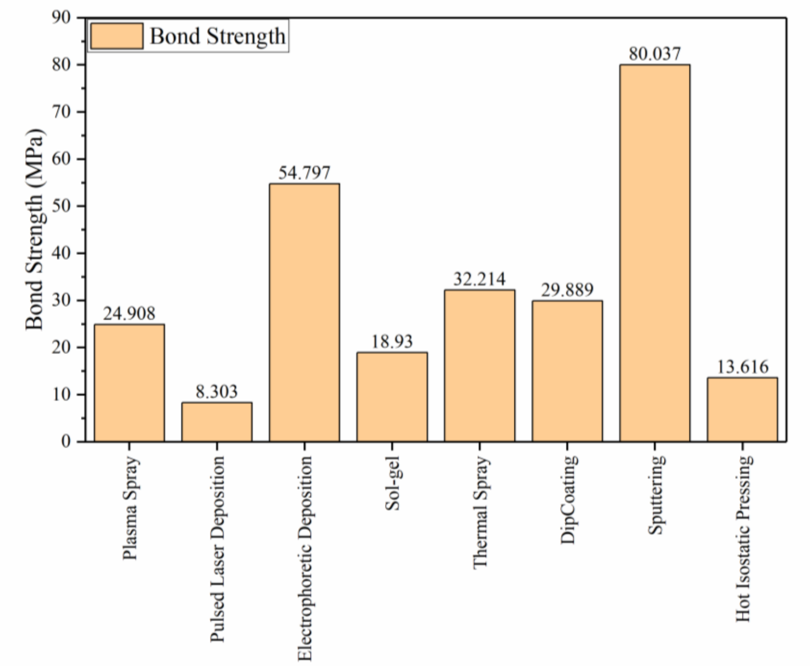


Fig. 5. Bond strength of hydroxyapatite coting over Ti_6Al_4V deposited by different coating technique. Based on data [37]

Coating technique also has a significant effect on the properties of coating. The effect of coating techniques on the bond strength of hydroxyapatite coated Ti_6Al_4V titanium alloy was studied by Mohseni et al. [37]. Bond strength of hydroxyapatite coting over Ti_6Al_4V deposited by different coating technique is shown in Fig. 5. It can be observed that the highest bond strength was obtained for hydroxyapatite coated by sputtering technique. On the other hand, the lowest bond strength was observed for pulsed laser deposited hydroxyapatite coating over Ti_6Al_4V substrate.

Contact angle. Contact angle is the measure of wettability of a coating material. Low contact angle (less than 90 °) signifies the hydrophilic nature of coating, while high contact angle (more than 90 °) signifies the hydrophobic nature of coating. High wettability (low contact angle) is desired for a coating material because wettability increase the proliferation, cell attachment, and mutual interaction of cells. Variation in contact angle of Ti alloy with different coating materials is shown in Fig. 6.

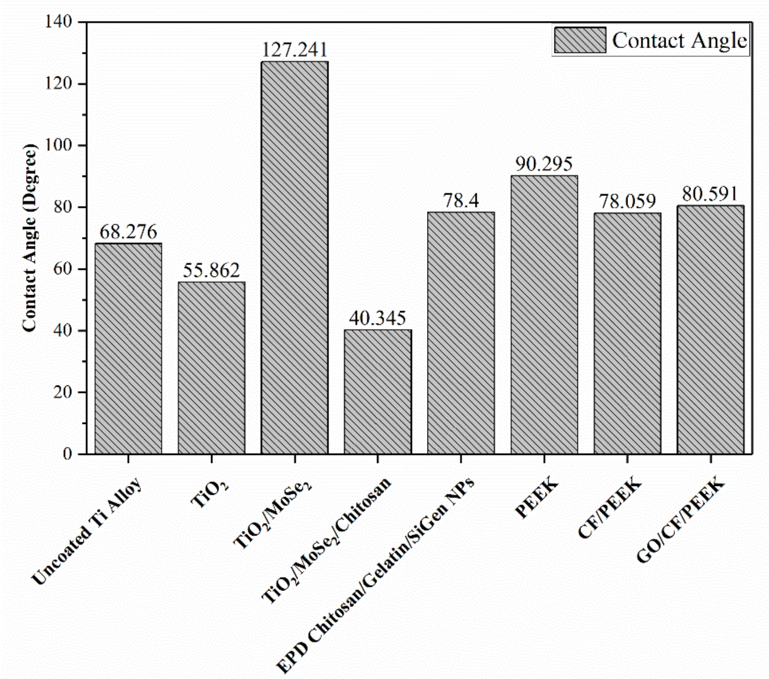
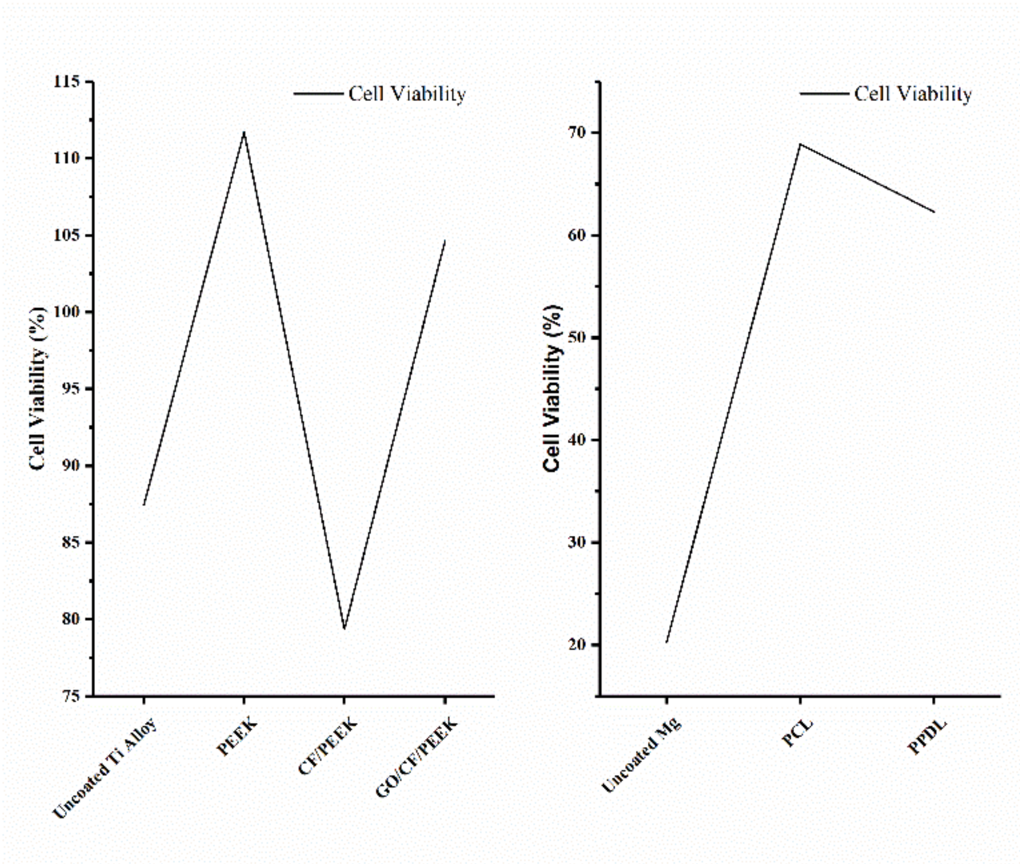


Fig. 6. Variation in contact angle of Ti alloy with different coating materials. Based on data [38–40]

Chai et al. [38] coated the titanium alloy with three different materials namely TiO_2, $TiO_2/MoSe_2$, and $TiO_2/MoSe_2$/Chitosan. It was observed that the lowest contact angle of 40.345 ° was obtained for $TiO_2/MoSe_2$/Chitosan coated sample. This indicates that $TiO_2/MoSe_2$/Chitosan enhance the wettability and will be helpful in combating infections. Aydemir et al. [39] investigated the effect of bioactive glass (Chitosan/Gelatin/SiGenNPs) composite coating on the contact angle of titanium alloy coated by electrophoretic deposition (EPD) technique. It was observed that the contact angle increased slightly. And hence the wettability reduced. Similar effect of polymer and composite coatings i.e., PEEK, carbon-fibre/PEEK (CF/PEEK), and graphene oxide/carbon-fibre/PEEK (GO/CF/PEEK) on the contact angle of coated titanium alloy was observed by Qin et al. [40].

Cell viability. Cell viability is the measure of biocompatibility of an implant material. If cell viability is high, then the fracture of living and healthy cells in the population is high. So, high value of cell viability is desirable. Hence, the coating materials that increase the cell viability will be preferred. Variation in cell viability of Ti and Mg alloys with different coating materials is shown in Fig. 7. Qin et al. [40] investigated the biocompatibility of titanium alloy coated with polymer (PEEK) and composites (CF/PEEK and GO/CF/PEEK). It was observed that the highest cell viability was observed for PEEK coated titanium alloy. Hence, the PEEK coated titanium alloy was most biocompatible. But, the wettability was low for PEEK coated titanium alloy (Fig.6).



(a) (b)

Fig. 7. Variation in cell viability of Ti alloy (a) and Mg (b) with different coating materials. Based on data [40-42]

For GO/CF/PEEK coated titanium alloy, both the wettability and the cell viability were high. Hence, the composite GO/CF/PEEK coating is preferable for titanium alloys. Ritwik et al. also investigated the biocompatibility of titanium alloy coated with chitosan [41]. It was observed that the cell viability enhanced after the chitosan coating over the titanium substrate.

Mahapatro et al. investigated the effect of polymer coatings on the biocompatibility of coated magnesium alloys [42]. It was observed that the cell viability was increased after the coatings of poly-caprolactone (PCL) and poly-pentadecalactone (PPDL) on the magnesium alloy. Highest cell viability was observed for the magnesium substrate coated with PCL. Hence, PCL is a better coating material for magnesium alloy if high biocompatibility is desired.

Surface roughness. Surface roughness is also one of the important property of an implant material. If surface roughness is high, tissues growth in the cavities will be high [43]. High tissue growth in cavities enhance the bonding between the implant and tissue. So, high surface roughness is desirable. Variation in surface roughness of steel and Ti alloy with different coating materials is shown in Fig. 8.

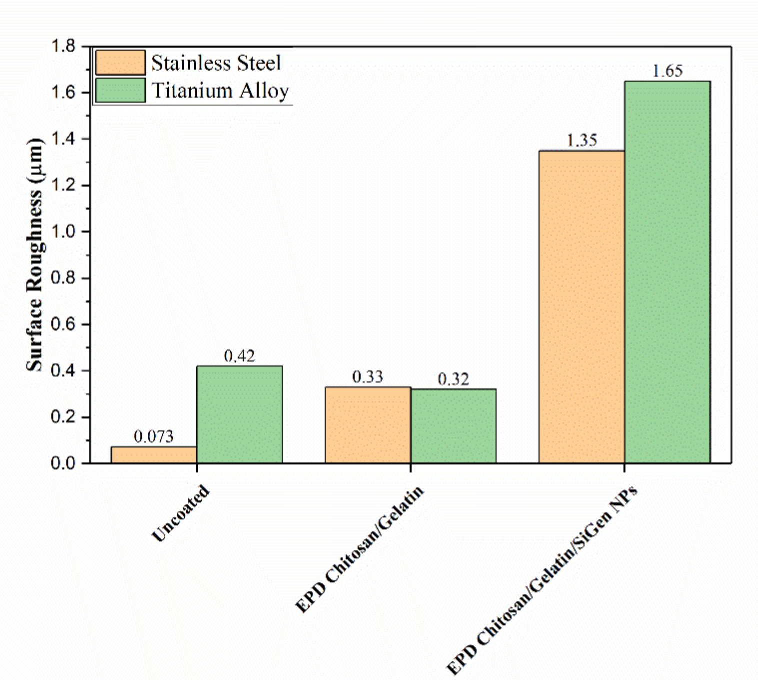


Fig. 8. Variation in surface roughness of steel and Ti alloy with different coating materials. Based on data [39,43]

It can be observed that the surface roughness increased after the composite coatings of chitosan/gelatin and chitosan/gelatin/SiGenNPs over the stainless steel substrate [39]. On the other hand, coating of chitosan/gelatin over titanium alloy slightly reduced the surface roughness. Surface roughness of chitosan/gelatin/SiGenNPs coated titanium sample was highest. Hence, chitosan/gelatin/SiGenNPs composite coating deposited by electrophoretic deposition technique is useful for both titanium alloys and stainless steel implants.

Oxide formation. Oxide formation is an important property for a metallic coating material. Metallic oxides are generally very less reactive and biocompatible in nature. Hence, the high oxide formation at the surface is desirable for an implant material.

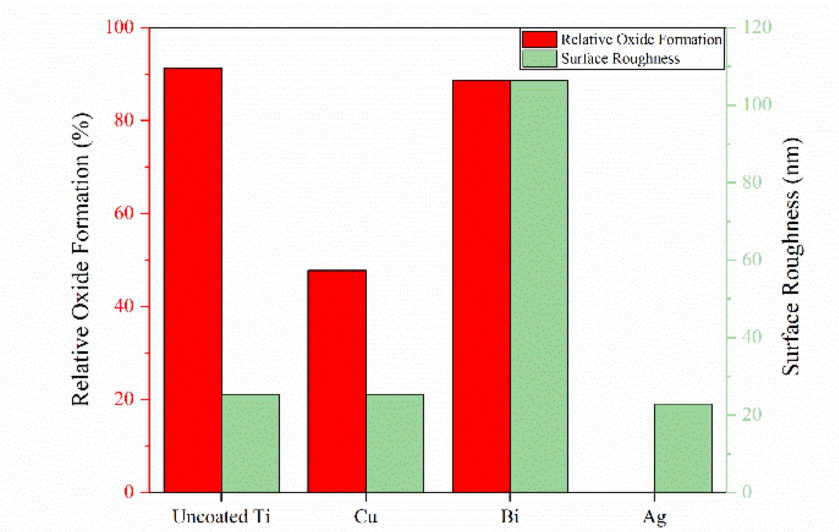


Fig. 9. Variation in oxide formation and surface roughness of Ti alloy with different coating materials. Based on data [44]

Gosau et al. investigated the oxide formation and surface roughness of coated and uncoated titanium implant materials [44]. Variation in oxide formation and surface roughness of Ti alloy with different coating materials is shown in Fig. 9. It can be observed that for the uncoated titanium sample, oxidation was high, but surface roughness was low. Bi coating enhanced the surface roughness and oxide formation was also very high (around 89 %). On the other hand, oxide formation was negligible in Ag coated sample because of very low reactivity of Ag. Oxide formation was around 50 % in Cu coated sample. Hence, the Bi is a very suitable coating material for titanium alloy implants.

Hardness. Surface hardness of an implant is dependent on the coating material. Higher the hardness, lower will be wear rate. So, the addition of hard coating over implant, reduces its wear and scratch and enhance its life. The effect of coating materials on the surface hardness of implants was studied by Vladescu et al. [45]. Surface hardness of implants with different coating materials is shown in Fig. 10. It can be observed that the highest hardness was obtained for an implant coated with diamond like carbon (DLC).

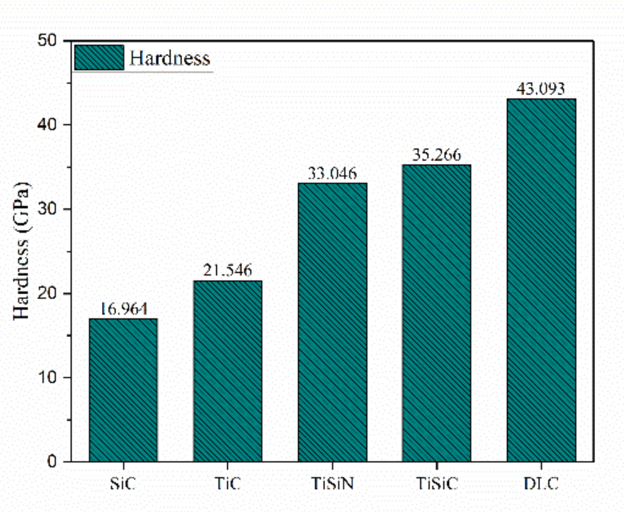


Fig. 10. Surface hardness of implants with different coating materials. Based on data [45]

Effect of coating thickness and processing parameters on bond strength

Coating thickness and processing parameters also have significant effect on the bond strength. Effect of these parameters on the bond strength is studied in detail in the following subsections.

Effect of coating thickness. Coating thickness is having a significant effect on the properties of a coated material. As an implant, it is used inside the body. So, it will work inside a corrosive environment. In a corrosive environment, materials dissolve at a higher rate. Hence, the thickness of a coating should be sufficient that it will last for the intended working life of the implant material. Coating thickness also affects other properties such as cell viability, corrosion resistance, surface roughness, and bond strength. Effects of coating thickness on the above-mentioned properties are studied in the following sections.

Kuo et al. investigated the effect of parylene coating thickness on the bond strength, cell viability, and surface roughness of the NiTi alloy [46]. Variation in bond strength, cell viability, and surface roughness with coating thickness is shown in Fig. 11,12. It can be observed that the cell viability decreased with an increase in the coating thickness. On the other hand, bond strength increased with an increase in the coating thickness. Surface roughness also increased initially with an increase in the coating thickness. On further increasing the coating thickness (beyond 5 μm), surface roughness decreased [46].

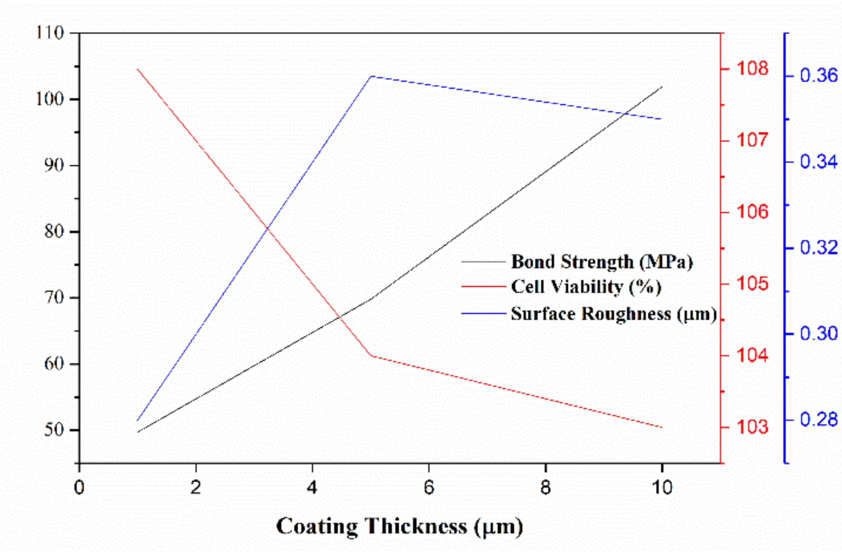


Fig. 11. Variation in bond strength, cell viability, and surface roughness with coating thickness. Based on data [42,46]

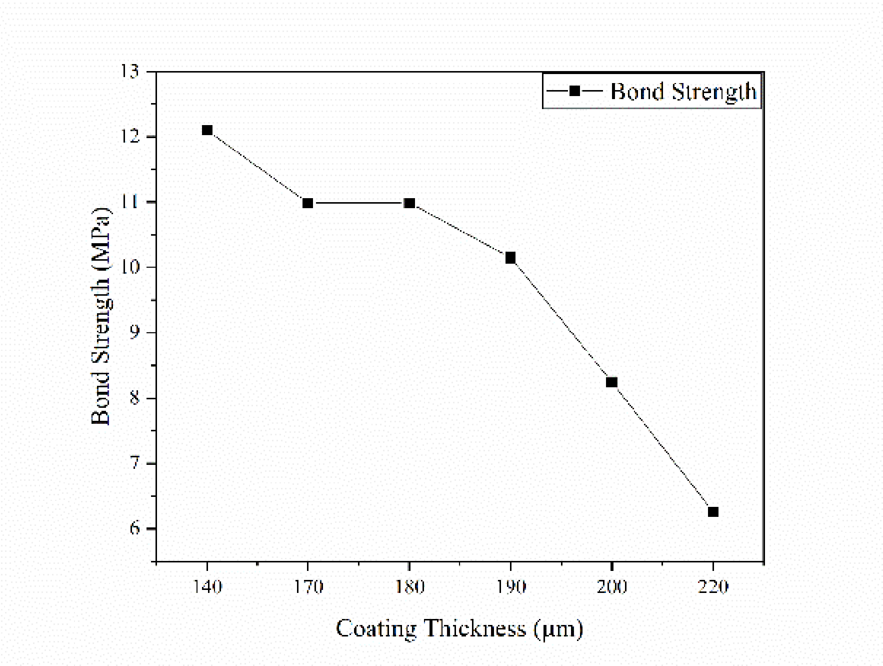


Fig. 12. Variation in bond strength with coating thickness. Based on data [37]

Mohapatro et al. [42] investigated the effect of coating thickness on the corrosion rate of coated magnesium substrate. It was observed that the corrosion rate reduced tremendously after the coating of PCL, as shown in Fig. 13. On increasing the coating thickness, corrosion rate decreased significantly.

Mohseni et al. [37] investigated the effect of hydroxyapatite coating thickness on the bond strength of Ti_6Al_4V titanium alloy. Variation in bond strength with coating thickness is shown in Fig. 12. It can be observed that the bond strength decreased with an increase in the coating thickness. So, the results are contrary to the results obtained by Kuo et al. [46]. This can be due to poor cohesion and adhesion of hydroxyapatite.

So, coating thickness has a mixed effect on the bond strength, cell viability, corrosion rate, and surface roughness. Corrosion resistance and bond strength increased with an increase in the coating thickness. On the other hand, cell viability decreased. So, selection of coating should be based on the required application.

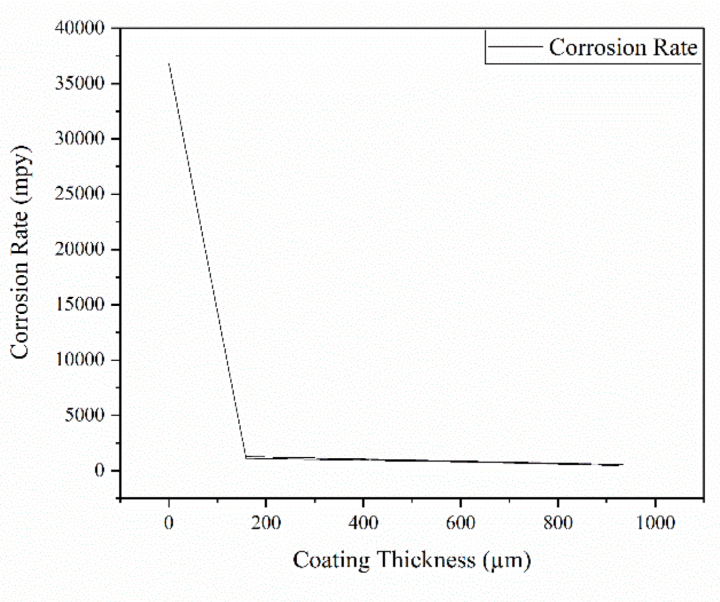


Fig. 13. Variation in corrosion rate with coating thickness. Based on data [37]

Effect of hot isostatic pressing temperature. Hot isostatic pressing is a technique used for the densification of the powders. This technique can be used for improving the bond strength of coating on the implant. Mohseni et al. investigated the effect of the temperature of hot isostatic pressing on the bond strength of hydroxyapatite coated Ti_6Al_4V titanium alloy [37]. Variation in bond strength with hot isostatic pressing temperature is shown in Fig. 14. It can be observed that the bond strength increased with an increase in the temperature of hot isostatic pressing for hydroxyapatite coating of 160 µm. On the other hand, bond strength decreased with an increase in the temperature of hot isostatic pressing for hydroxyapatite coating of 200 µm. So, the thicker coating has adverse effects on the bond strength.

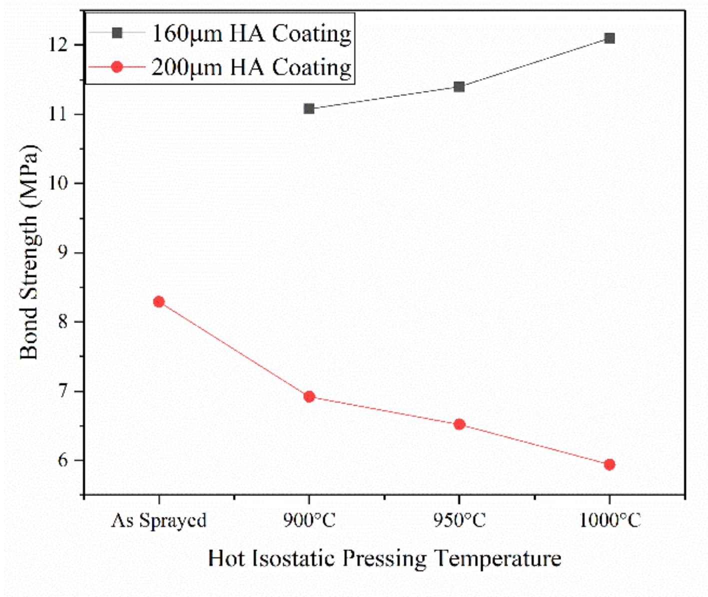


Fig. 14. Variation in bond strength with hot isostatic pressing temperature. Based on data [37]

Effect of surface cleaning and treatment. Surface of substrate play an important role in the coating efficiency. If the surface is not clean, coating adhesion will be poor. Also, various surface treatments and modification enhance the coating bond strength. Some of the surface treatment techniques are shown in Fig. 15. Hsiung et al. [47] investigated the effect of cleaning and cryogenic treatment on the bond strength of the hydroxyapatite coated Ti_6Al_4V implant material. Bond strength of untreated and cryogenically treated hydroxyapatite coated Ti_6Al_4V implant material cleaned by two different processes is shown in Table 2. It can be observed that the bond strength of ultrasonically cleaned sample was more than the bond strength of the sample cleaned by high pressure air. It shows that the ultrasonic cleaning is better than the high pressure air cleaning. Also, bond strength of cryogenically treated samples was higher than the bond strength of the untreated samples. So, the cryogenic treatment is a very useful technique to enhance the coating efficiency.

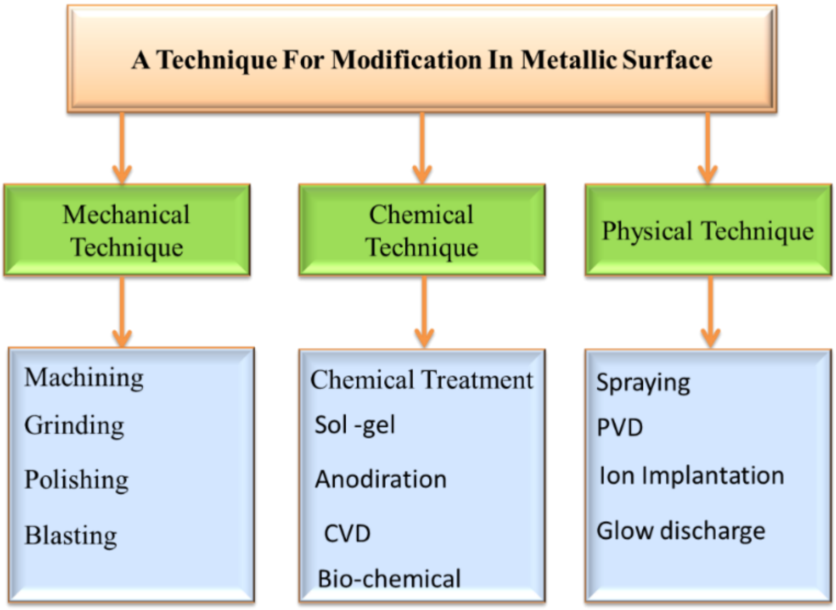


Fig. 15. Surface modification/treatment techniques

Table 2. Bond strength of untreated and cryogenically treated hydroxyapatite coated Ti6Al4V implant material

	Bond Strength, MPa	
	Ultrasonic Cleaning	Pressurised Air
Untreated	26.56	18.91
Cryogenic Treatment	36.65	29.30

Conclusions

After detailed literature survey of coated and uncoated implant materials, following conclusions have been drawn:

1. It was observed that the ceramic, polymer, metallic, and composite are widely used for coating metallic implants;
2. Biocompatibility of polymer coatings were relatively less in comparison to ceramic and composite coatings. Also, wettability of polymer coatings was less;
3. On the other hand, ceramic coatings were highly biocompatible and wettability was also high;
4. For metallic coating, biocompatibility was less because of high reactivity. But, metals (like Bi) that form protective oxide layer on the surface of implants exhibits very good biocompatibility and mechanical strength;

5. Surface treatment is a very good technique to enhance the adhesion between the coating and the substrate;
6. Coating technique also significantly affect the properties of coating. Highest bond strength is obtained by sputtering coating.

Future Scope
Comparative study on the mechanical properties of nano-coated materials can be done in order to identify stress, elongation, and swelling behaviour. In this work, the effect of coatings on only cell viability, contact angle, surface roughness, and corrosion rate is studied. Other biological and mechanical properties can be investigated.

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