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REVIEW PAPER

Corrosion Features of Titanium Alloys in Dental Implants: A Systematic Review

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Abstract

Background Titanium and its alloys are the gold standard for dental implants because of their unique combination of chemical, physical, and biological properties, and in particular thanks to their biocompatibility, resistance to corrosion, and mechanical properties. However, despite advanced technologies to avoid corrosion of dental implants, the mechanisms toward the release of metals and their role in the onset of peri-implant diseases are still under-investigated. The combination of stress, corrosion, and bacteria contributes to implant failure. Recent studies suggest the existence of wear/corrosion products may correlate with peri-implantitis progress by triggering microbial dysbiosis, the release of pro-inflammatory cytokines, and animal bone resorption.

Aim The aim of the present article is to highlight the various features of corrosion of titanium alloys used in dental titanium implants, to review the evidence toward biocorrosion in the oral environment and to discuss the methodological and electrochemical aspects of surface treatments and titanium-based alloys.

Methods The present review was conducted according to the Preferred Reporting Items for Systematic reviews and Meta-Analyses extension for Scoping Reviews (PRISMA-ScR) guidelines. The research question was about the corrosion of titanium alloys used in dental implants and their consequent biological side effects on peri-implant tissue.

Results After screening the duplicates and following the application of exclusion criteria, the full texts of 23 articles were included in the review.

Conclusions The development of improved strategies toward the reduction of corrosion and degradation of titanium alloys used for dental implants is crucial, also to prevent metal release in the tissue surrounding them to prolong their lifetime. As chemical and physical properties are crucial for the electrochemical behavior of the implant material, the development of appropriate alloys or coatings/layers for corrosion inhibition is mandatory.

Keywords Titanium alloys · Dental implant · Corrosion · Titanium · Oral

Introduction

Cobalt-based alloys, magnesium (Mg), and its alloys have proven their applicability as bone implants. However, titanium (Ti) is the most popular material used for dental implants, and its alloys have been widely applied clinically. An ideal implant should be biocompatible, possess high strength, fatigue and fracture toughness behavior and should be able to withstand the reactive environment it is exposed to inside the human body [1–96].

Ti-based dental implants have become a predictable standard of care for replacing missing teeth, thanks to the good mechanical properties, the resistance to corrosion, and the excellent biological performance of Ti, that is able to produce a spontaneous Ti oxide layer after exposure to oxygen atmosphere (mainly TiO_2) [2, 4, 7, 10–15]. However, corrosion of dental implants may jeopardize the mechanical

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stability of the device, as well as the integrity of the surrounding tissue. Pure titanium, in spite of its corrosion-resistant characteristics within controlled environments and in the absence of load, it can corrode under oral conditions and in association with cyclic loads, thus influencing the mechanical stability of dental implants. In fact, exposure to acidic substances and microbial metabolites can lead to a reduction in pH, potentially causing the rupture of the passive film. Moreover, the TiO_2 barrier might have a poor tribological efficacy and it might be removed/disrupted under loading [1–30]. Metallic debris may induce an inflammatory response. In fact, the products yielded by corrosion may have cytotoxic effects on the tissue surrounding the implant [1–37].

Types of corrosion associated with metallic implants may be galvanic, fretting, pitting, and crevice corrosion. As for galvanic corrosion, theoretically, titanium screw would represent the anode, the metallic fill the cathode, and saliva would be the electrolyte. Fretting corrosion would occur because of disruption of the protective layer on titanium screws, while pitting corrosion would be the consequence of the spontaneous breakdown of the passivating film on a flat or overexposed area. Finally, crevice corrosion is associated with uneven surfaces.

These corrosive factors, together with wear induced by implantation procedures (e.g., friction, micro-motion), may determine an undesired release of metallic ions and particles from the implant to the surrounding tissues, thus eventually leading to severe biological complications such as peri-implant diseases [1–8, 13–16, 25, 36–63]. Furthermore, the rate of a corrosion process depends on the oxide layer formed, pH, the concentration and composition of the electrolyte, and the transport of oxygen vacancy across the film.

Therefore, in consideration of corrosion as a potential risk factor for peri-implantitis, the knowledge of the influence of corrosion-induced release of ions and particles as a driving factor for peri-implant diseases and early/late failure of dental implants is fundamental and clinically relevant.

The aim of the present article is to review the currently available information about the corrosion of titanium alloys used in dental implants and their consequent biological side effects on peri-implant tissue.

Materials and Methods

The present review was conducted according to the Preferred Reporting Items for Systematic reviews and Meta-Analyses extension for Scoping Reviews (PRISMA-ScR) guidelines. The research question was about the corrosion of titanium alloys used in dental implants and their consequent biological side effects on peri-implant tissue. A literature search was conducted in four electronic databases (Medline/PubMed, Scopus, Embase, and Web of Science) using the combination of the terms "dental implants" OR "surface treatments" OR "alloys" AND "corrosion" OR "electrochemical" OR "degradation.

Articles not in English language were excluded.

The following types of articles were excluded: duplicated/overlapping articles, animal studies, conference proceedings, expert statements, editorials, case reports, and nonoriginal papers.

Data sharing not applicable to this article as no datasets were generated or analyzed during the current study.

Results

The initial search resulted in 543 articles. After screening the duplicates and following the application of exclusion criteria, the full texts of 23 articles were included in the review (Fig. 1).

Discussion

After dental implants are inserted in the jaws, the integrity of the protective TiO2 passive layer, as well as the maintenance of their physicochemical properties, depends on the hostile electrolytic oral environment. In fact, oral fluids (saliva, blood plasma) are characterized by organic and inorganic substances in combination with a pH between 6 and 7, that may be reduced by some factors such as microbial metabolites and corrosive substances, thus making the environment acidic and highly reactive to chemically attack the metallic surfaces [20–25, 36, 42, 62, 70–74, 82].

Cl^-, F^-, H^+, and other corrosive substances, may be found not only in saliva, but also in prophylactic commercial formulations, such as toothpaste and mouth rinses, and foods.

In particular, in literature, the F^- ion concentration adversely affects the corrosion resistance of titanium-based materials because of the formation of hydrofluoric acid (HF) from fluoride ions when the aqueous environment is acidified by some food or microbial metabolites. HF is strongly reactive to metals because of its chemical characteristics prone to induce the breakdown of the TiO2 protective layer ($TiO_2 + 4HF \rightarrow TiF_4 + 2H_2O$). Therefore, HF not only promotes the degradation but also the discoloration of the metal surface because of the generated fluoride-titanium compounds [53–69].

One of the significant factors contributing to the reduction in the corrosion resistance of titanium is the action of oral bacteria. When dental implants are inserted in the oral cavity, the outer implant surface and the micro gaps between the implant and abutments are covered by protein-rich fluid

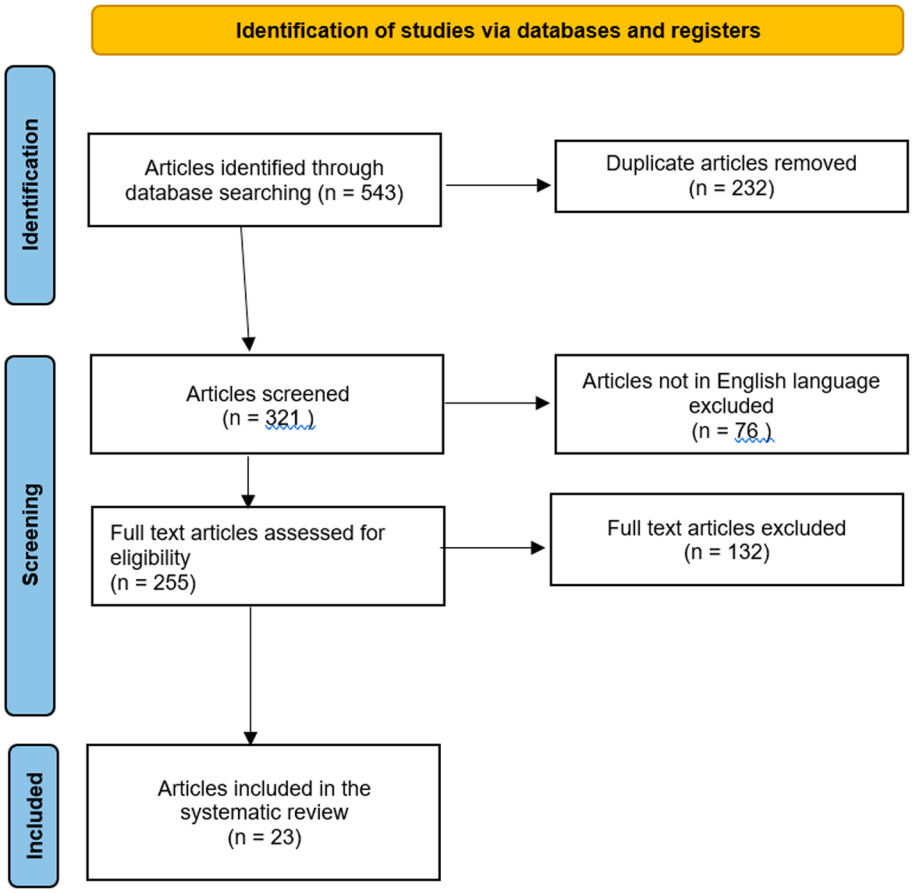

Fig. 1 PRISMA flow chart

pellicle (saliva, blood), thus promoting the adhesion of bacteria to these surfaces. Bacterial acidic metabolites and oxygen level deficiency may determine oxidation reactions between the biofilm-covered Ti surface and the exposed Ti, increasing the corrosion rate of the implant material [20–41].

Therefore, bacterial cells and physical, chemical, microbiological, and inflammatory corrosion processes promote the surface damage of dental implants and contribute to the implant surface degradation. This may adversely alter the microenvironment conditions of peri-implant tissues, leading to cytotoxic and inflammatory reactions. Finally, such events compromise the success of dental implant rehabilitations [10–32].

The processes involved in the corrosion of titanium alloys of dental implants may be mechanical (wear particles/debris), electrochemical (corrosion-related free metal ions, organometallic complexes, and salts), and/or a combination of both mechanical and electrochemical processes (tribocorrosion).

Tribocorrosion occurs under the dual action of wear and corrosion under a variety of conditions such as sliding, fretting, rolling, impingement in a corrosive medium.

The most common types of electrochemical corrosion found in titanium alloys used for implant applications are galvanic, fretting, and pitting/crevice corrosion, as well as environmentally induced cracking (EIC) [20–25, 36, 42, 62, 70–74, 82].

Galvanic corrosion is associated with a direct contact of two dissimilar metals in an electrolytic solution. The difference in electrochemical potential of the two metals promotes oxidation of the more reactive metal. This becomes the anode, which generates a flow of electrons and ions to the cathode. Galvanic corrosion is not frequent in dental implant applications because of the presence of only one component, the dental screw, and the insulating nature of the protective passive layer that forms on the surface. Anyway, in some cases, the surrounding tissue might behave as a medium for electrical flow between metallic implants and other types of alloys used in dentistry for amalgams or orthodontic devices [1–15].

Fretting corrosion is due to the repeated micro-motion or friction of a metal component against another material that causes mechanical wear and breaks up the passivating layer on the contact surface of the metallic device. Fretting between dental implants and bone during implantation and due to cyclic loads imparted from chewing has been suggested as a cause of Ti corrosion and metal ion release. The release of metal debris and ions has been linked to inhibition

of cell differentiation, phagocytosis of Ti particles by macrophages and other cells, and inflammation. Abnormal electrical signals may affect the stability of the adjacent tissue, and fretting corrosion may amplify other types of corrosion by rupturing the passivating film and exposing bare Ti [10–23].

Pitting corrosion (i.e. the result of the spontaneous breakdown of the passive film on a flat and evenly exposed area) is not likely to occur on Ti surfaces.

Instead, crevice corrosion has been observed on Ti and Ti alloys, and it consists of a localized corrosion due to a geometric confinement in the design of the device or from a previously corroded region on the surface [26–31].

Finally, EIC is the most common cause of corrosion in implants for bone applications, and because of its localized nature, may go unnoticed until failure of the implant. It consists of the brittle mechanical failure of metallic devices under stress levels significantly lower than their ultimate tensile strength. The magnitudes of the forces that can cause EIC vary over a wide range and include forces that, under non-corrosive conditions, would be considered negligible [10–23].

Because of these processes, a series of biochemical reactions might be triggered at the biointerface microenvironment.

As a consequence of the corrosion processes of dental implants and the Ti particle/ion release, signaling factors promoting the differentiation and recruitment of osteoclast cells responsible for peri-implant bone resorption determine an induction of inflammatory process in the peri-implant soft tissue cells and bone cells [10–23, 41–52].

Once metallic particles/ions are released, the peri-implant inflammatory process may be caused by the activation of the function of phagocytic cells such as neutrophils and macrophages, by the stimulation of communication pathways of osteoblastic cells, or by the promotion of microbial accumulation in the degraded rougher surface region. In particular, the phagocytosis of Ti ions may determine a higher expression level of pro-inflammatory cytokines (e.g., *Interleukin* 1β, *Interleukin* 6, and Tumor necrosis factorβ). Finally, the induction of receptor activator of nuclear factor kappa-*B* ligand (RANKL) expression within osteogenic cells is promoted, thus indirectly promoting RANKL-induced osteoclast differentiation and consequent tissue inflammation and bone resorption [41–52].

The cytotoxic effect of Ti products on the inflammatory response of human cells has been demonstrated for particles and Ti ions.

In the literature, evidence for Ti degradation in diseased peri-implant tissues has been detected by inductively coupled plasma mass spectrometry in submucosal plaque, soft-tissue biopsies, and exfoliative cytologic samples in greater amount in diseased peri-implant mucosa than in healthy sites. Nevertheless, for both healthy and inflamed tissue biopsies, the Ti concentration found (7.3 to 38.9 μM) was within the levels needed to activate the IL-1β secretion from human macrophage in vitro, that is a phenomenon closely related to stimulating an in vivo proinflammatory reaction.

Therefore, there seems to be poor specificity between the biological impact of Ti concentration and the pathological process of peri-implant diseases [62–74].

To resume, it is likely a multidirectional pathway loop for the degradation of Ti surface in the oral environment: (1) first of all, wear, acidic substances, and metabolites released from oral bacteria promote the degradation of the peri-implant microenvironment, thus provoking the passive oxide layer breakdown and consequent pitting and galvanic attacks; (2) then, the corroded Ti surface with an increased roughness provides additional niches for bacterial recolonization; (3) the microbial accumulation promotes oxygen level deficiency that prevents the re-formation of the passive oxide layer; (4) finally, corrosion products induce microbial dysbiosis, the occurrence of inflammatory reaction, and the consequent generation of acid products (hydrogen peroxide and H^+) that in the end also negatively affect the corrosion resistance of Ti [62–74].

An interdisciplinary engineering and biomedical approach is needed to improve the strategies to reduce the corrosion of dental implants and the consequent undesired effects.

Exploring Titanium Alloy Compositions for Dental Implants

Altering the composition of titanium alloys for dental implants to resist corrosion is the most straightforward option with the aim of creating a material that can achieve passivity, regulate the hydrogen evolution reaction, and reduce the anodic/cathodic activity directly. In fact, several elements are acknowledged to inhibit the degradation process by leading the growth of highly stable passive oxide films and microstructures [62–74, 76–96].

The main elements proposed for titanium alloys and their role in improving their electrochemical properties are resumed in Table 1. When an alloying element is added to Ti, the alloy may undergo phase transformation reactions, that may result in three microstructure phases (β, $\beta+\beta$, and β) with intermetallic variants. Anyway, there is no consensus regarding the best crystalline phase to prevent corrosion: β crystalline phase is expected to be more resistant to dissolution and stabler than the β-phase, but the single β-phase in Ti alloys has proven a better electrochemical behavior than $\beta+\beta$ and β alloys.

Zirconium (Zr), tantalum (Ta), niobium (Nb), chromium (Cr), and molybdenum (Mo) are normally added to Ti to

Table 1 Main elements proposed for titanium alloys and their role in improving their electrochemical properties

Alloying element	Atomic number	Classification	Effect on Ti alloy microstructure and electrochemical parameters
Zr	40	Transition metal	Zr addition determines higher polarization resistance (*R*p) and lower values of corrosion current density (*i*corr), corrosion rate, and capacitance parameters of Ti–Zr alloy
Ta	73	Transition metal	Ta incorporation forms a stable Ta_2O_5 passive film and a β phase in the Ti matrix, that decreases pitting initiations, the corrosion rate, and *i*corr while enhancing the corrosion potential (*E*corr) parameter of Ti–Ta alloys
Nb	41	Transition metal	Nb addition decreases the corrosion rate, *i*corr, and passivation corrosion density (*i*pass) values, while increasing the *E*corr parameter of Ti–Nb alloys
Cu	29	Transition metal	Cu addition determines nobler *E*corr and higher *R*p, while the *i*corr decreases with increasing Cu content
Ag	47	Transition metal	Ag addition to create Ti–Ag alloys increase *E*corr, while decreases *i*corr
Pd	46	Transition metal	Pd addition accelerates the protective cathodic reactions and inhibits the dissolution of Ti while decreasing *i*corr and capacitance and enhancing the *R*p of Ti-0.2Pd alloy
Cr	24	Transition metal	Cr addition forms a Cr-rich oxide film that improves the *R*p and diminishes the *i*corr of the Ti–Cr alloy
Mn	25	Transition metal	Mn addition enhances the cathodic reaction but does not improve the corrosion resistance of Ti considerably
Co	27	Transition metal	Ti–Co alloys show similar corrosion behavior to pure Ti
Mo	42	Transition metal	Mo addition improves the stability of the anodic oxides, increasing *R*p and decreasing *i*corr and *i*pass as the content of Mo enhances in Ti–Mo alloys
Fe	26	Transition metal	Fe addition may reduce the *i*corr and increase the critical pitting potential (*E*pit) and *R*p parameters of Ti–Fe alloys
Bi	83	Post-transition metal	Bi addition slightly increases *E*corr values but shows a significantly lower *i*corr in an electrolyte containing fluoride and acid lactic
In	49	Post-transition metal	In addition decreases the corrosion rate and *i*corr, enhancing *R*p values of Ti–In alloy
Mg	12	Alkaline earth metal	Mg addition decreases the *E*corr,while increasing the *i*corr

form a stable and resistant oxide film when in contact with the environment.

Ta incorporation forms a stable Ta_2O_5 passive film and a β phase in the Ti matrix, which reduces pitting initiations, the corrosion rate, and *i*corr while enhancing the corrosion potential (*E*corr) parameter of Ti–Ta alloys [76–96].

Instead, adding Mo to pure Ti improves the stability of the anodic oxides, increasing *R*p and decreasing *i*corr and *i*pass as the content of Mo enhances in Ti–Mo alloys.

As for intermetallic compounds, they will dissolve preferentially on the surface by developing a galvanic cell with the matrix, which impairs the alloy's electrochemical stability. Single-phase alloys exhibit better electrochemical properties. When more than one phase and/or diverse crystallographic orientations are present, it is necessary to achieve grain refinement and an even distribution of the elements.

In fact, a fine microstructure determines an "enveloping effect" by modifying the cathode/anode area ratio between the intermetallic and the matrix to minimize the galvanic effects and provide corrosion protection of the less noble phase, while homogeneous microstructures and greater elemental distributions may cause an increased corrosion resistance because of the improved stability and durability of the passivation films formed on the matrices.

Table 1 only resumes binary Ti alloys, but several other elements have been used to develop corrosion resistant ternary, high entropy alloys (HEAs), or compositionally complex alloys (CCAs). For this reason, researchers should first model combinations of elements by computational tools that consider first-principles calculations to predict material properties and electrochemical mechanisms before testing it by in vitro studies.

Surface Treatment Options for Dental Implants

Surface treatments are applied to the implant substrate to minimize the corrosion damages by avoiding the penetration of corrodents on metal underneath and preventing the electrochemical reactions [16–23, 45–96]. The outcomes of the most frequent technologies proposed to protect against corrosion of dental implants surfaces are resumed in Table 2.

The electrochemical stability of coatings is influenced by their elemental and crystalline composition. Moreover, the reinforcement of the oxide layer with homogeneously distributed functional and stable compounds and crystal phases by coating/film deposition techniques has demonstrated to enhance the corrosion performance of the implant material.

Table 2 Outcomes of the most frequently used surface treatment technologies proposed to protect against corrosion of dental implants surfaces

Surface treatment technology	Outcomes related to corrosion behavior
Grit blasting	Some particles can leach from the surface, impairing the electrochemical stability of the material and determining an irregular topography that may disrupt oxide films, thus reducing corrosion resistance
Acid etching	The formation of a TiH intermediate layer by the acid reaction with the Ti substrate positively affects the electrochemical stability, allowing the growth of a new stable oxide layer
Sandblasting/acid etching	In spite of the irregular topography that may decrease the corrosion resistance, the etching process cleans any remaining impurities from blasting and creates a TiH intermediate layer that may contribute to protect the surface against corrosion
Sol–gel	The formation of dense and crack-free coatings with adequate adhesion strength to the substrate may create a protective barrier effect, thus causing a reduction in the corrosion rate. Functional compounds can be added to the layer, thus providing additional corrosion resistance
Anodic spark deposition or plasma electrolytic oxidation	This technique thickens the oxide layer that behaves as a barrier to ion diffusion. Therefore, it leads to the growth of a fully oxidized protective surface with different microstructures and compositions that may contribute to avoiding the electrochemical dissolution of the implant at the metal-electrolyte interface
Electrophoretic deposition	It aims to create a compact and uniform coating that causes a reduction of the penetration rate of the solution into the coating. Functional compounds can be incorporated to improve the local corrosion resistance of the coating matrix
Physical vapor deposition (magnetron sputtering)	It allows the creation of a more stable, compact, and homogeneous film than the natural oxide layer, in order to decrease the penetration of ionic species through the film, and to mitigate the electrochemical degradation process
Chemical vapor deposition	It allows the formation of a dense and thin film that acts as a physical diffusion barrier blocking the charge transfers between the substrate surface and the electrolyte effectively
Plasma spraying	This method, in spite of forming a coating that may present several defects (such as pits, voids, microcracks, and pores) that are detrimental to electrochemical stability, may involve reinforcement compounds to compensate for these disadvantages, enhancing the corrosion resistance
Hydrothermal- and alkali-based treatment	It aims to obtain a dense and large coating thickness to physically isolate the substrate from the corrosive fluid, thus preventing its penetration into the coating underneath and providing effective initial protection
Ion implantation	This technique aims to improve the corrosion resistance by changes in the crystallinity of the microstructure or alterations in the oxide composition of the passive film
Polyelectrolyte multilayers	This method aims to improve the corrosion behavior by the creation of multilayers that reduce the electrolyte permeability due to the strong ionic pairing between polyelectrolytes in adjacent layers, and that may also display an intrinsic self-healing behavior because of the introduction of nano-reservoirs or nano-reactors within the multilayer structure

To this aim, a series of mechanism and ideal criteria that coatings and films need to meet for corrosion inhibition have been proposed: the formation of stabler oxide films (for example containing TiO_2, ZrO_2, Nb_2O_5, Ta_2O_5) contribute to prevent the internal dissolution of the coating due to corrosive attacks; dense, compact, and defect-free layers should fill the substrate porosities in order to prevent the corrosive fluid to reach the surface of the implant; thick layers may decrease the dissolution of the coatings in the immersion medium; an improved ability of substrates to form passive layers after coating/film deposition determines a better protective behavior; a strong bond strength between the coating and the substrate might prevent the coating cracking and peeling off in the body fluid, thus avoiding local corrosion [16–23, 45–96].

Conclusions

The development of improved strategies toward the reduction of corrosion and degradation of titanium alloys used for dental implants is crucial, also to prevent metal release in the tissue surrounding them to prolong their lifetime.

As chemical and physical properties are crucial for the electrochemical behavior of the implant material, the

development of appropriate alloys or coatings/layers for corrosion inhibition is mandatory.

A thorough knowledge of corrosion mechanisms and the development of better methods to improve the corrosion resistance of dental implants may contribute to control peri-implant diseases and achieve safe and long-term dental implant rehabilitation treatment.

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