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Corrosion and degradation of metals and alloys in dentistry

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Abstract: Metal alloys have high strength and are competitive among other materials used in dentistry. The choice of a material for dental application, depends on several factors like corrosion behaviour, mechanical properties including strength and fabricability, cost, availability, biocompatibility, and aesthetic values. For metallic materials, the corrosion behaviour is the most important property to be considered, due to the biocompatibility and cytotoxicity of the products of the corrosion process. We will discuss the different types of corrosion of metals and alloys in dentistry and how to minimize it.

Keywords: *Dental corrosion, metals, alloys, chemical failure, degradation of metals.*

Introduction

Corrosion is defined as “the destruction or deterioration of a material because of reaction with its environment”. Also, “it is a chemical or electrochemical process through which a metal is attacked by natural agents, such as air and water, resulting in partial or complete dissolution, deterioration, or weakening.” [2, 3]. Corrosion always come as a consequence to tarnish, which is “a surface discoloration on a metal or a slight loss of the surface finish or gloss due to the formation of thin films, such as surface oxides, sulphides, or chlorides”. Fontana [3] and Jones [4] classified corrosion into eleven forms. Only nine forms occur in the oral environment.

Mechanism & Fundamentals of corrosion

Corrosion occurs by chemical or electrochemical means. Chemical corrosion or oxidation occurs when oxygen in the air reacts with metal without the presence of a liquid. Typically, chemical (dry) corrosion is not as damaging as electrochemical (wet) corrosion, but it is very sensitive to temperature. An electrochemical reaction involves metals, chemicals, and water that combine to form cells capable of generating electricity. The electrochemical cells contain four basic components: electrolyte, anode, cathode, and conductor. The electrolyte is the current carrying medium, such as water.

Definitions

1. Electrode potential: The difference in electric potential between an electrode and the [5]

2. High electrode potential (low dissolution tendency): If an element or compound has a positive standard electrode potential, it means it does not form ions as easily (be reduced and be an oxidizing agent). There is a greater tendency for reduction to occur.

3. Low electrode potential (High dissolution tendency): The more negative the value, the easier it is for that element or compound to form ions (be oxidised and be a reducing agent). There is a greater tendency for oxidation to occur.

4, Electrolyte: A medium that conducts electric current (through the movement of ions) as a result of a dissociation into positively and negatively charged particles called ions. This includes most soluble salts, acids, and bases dissolved in a polar solvent, such as water.

The anode (positive electrode) is in contact with the electrolyte, and corrosion will occur through an oxidation reaction that involves the formation of ions and the release of electrons. The cathode (negative electrode), no corrosion occurs because of a reduction reaction, which is the acceptance of electrons generated from the anode [5].

The conductor connects the anode and cathode to complete the circuit for current flow. In the wet corrosion process, all corrosion cells that are formed contain these four basic parts. Therefore, corrosion occurs from two simultaneous reactions: oxidation and reduction (redox) .

The most used metals naturally exist as compounds of oxides or sulphides, such as Fe3O4, Al2O3, or CuS. These compounds represent the thermodynamically stable form (lowest energy state).

Pure metals spontaneously react with oxygen, sulphur, or chlorine to revert to their lowest energy state, this is actually the process of corrosion. Gold and other noble metals are found in their pure form with lowest energy state, so they do not corrode [2].

Passivity

Passivity is defined as a “corrosion preventative mechanism where an oxidation layer forms a continuous film (passive layer) on a metal's surface that act as a barrier between the metal and the electrolyte, thus prevents further corrosion”. Active metals and alloys, lose their chemical reactivity and become extremely inert, because of the formation of a highly adherent and very thin oxide film on the metal surface, which serves as a protective barrier to further corrosion. It occurs in specific metals, such as Chromium, Aluminum, and Titanium [2, 5].

Re-passivation is repeating the process after a period of time due to the protective coating wearing down. The re-passivation potential is affected by the critical current density, pit current density and the thickness of the oxide film. The pit current density is “the current density below which pits stop growing”. While the critical current density is “the minimum current density needed to maintain the critical pit environment and prevent re-passivation”. The re-passivation potential decreases as thickness of the oxide film increases. Also, the re-passivation potential for a given metal film thickness increases when the electric current drops below the critical value of the pit current density. Similarly, the re-passivation potential will increase when the electric current is higher than the critical current density [30.]

Corrosion pathways

Three different electrolytic corrosive systems may exist in the oral cavity.

- The simplest cell consisted of a pure metal or single phase alloy electrode surrounded by tissue fluid and saliva.

- A cell with two electrodes that are physically separated, yet connected through saliva and tissue fluid may also occur in the mouth.

Both of these cells are unlikely to produce a high electron flow because of high resistance within the electrolyte.

- The electrodes are in direct contact or come into intermittent contact, this eliminates the high resistance of the two electrolyte systems and allows the possibility of a higher electron flow.

This is the electrolytic cell which is most likely to occur within a multiphasic alloy or where different alloys have been used to restore the same tooth. It is also likely to occur when different alloys come into the intermittent contact of mastication, when adjacent teeth restored with different alloys are in intimate contact, and When other alloys such as dental instruments or partial dentures contact a previously restored tooth.

Electrochemical potentials, current pathways, and resistances depend on whether there is no contact, intermittent contact, or continuous contact between alloy restorations [10].

a) Non-contacting Isolated Alloy Restoration

In case of a single metallic restoration, an eclectic current path between external surface exposed to saliva and interior surface exposed to dentinal fluids is formed.

Because the dentinal fluids contain a higher Cl concentration than saliva, so it is assumed the electrode potential of interior surface exposed to dentinal fluids is more active [11].

B) Nonisolated

For two restorations not in contact the extracellular fluid - saliva resistance, determines the extent to which the current will pass through. If the resistance is high, the current will not flow through the both. The interaction between the separated restorations will be minimized. Each restoration, though, will still generate its own current path loop [11].

Restorations Making Intermittent or Continuous Contact

a) Intermittent Contact

This occurs when two alloy restorations (one in the upper arch and the other in the lower arch) come into contact intermittently by biting. When the two restorations are in direct contact, a galvanic cell is generated with an associated galvanic current formed between the two restorations. The external current path can take a number of directions, with the least resistance path controlling. One entirely through extracellular fluids and the other partly through extracellular fluids and partly through saliva [11].

b) Continuous Contact

Another situation in which metallic restorations in the mouth are capable of generating galvanic currents involving two dissimilar metallic restorations in continuous contact, as in the combination of amalgam- gold alloy couples, between two amalgam restorations (one of conventional amalgam and the other a high-copper amalgam), two gold alloys with differences in noble metal content. Soldered appliances are also examples of dissimilar metals making continuous contact. Any multiphase microstructures are situations for galvanic corrosion to occur [11].

Different forms of corrosion

a. Chemical (dry) corrosion

Dry corrosion occurs in the absence of a liquid phase or above the dew point of the environment. In this case, a metal reacts with vapours and gases usually at high temperatures. These reactions can produce a tarnishing attack on exposed metals 15. In wet corrosion, a metal usually reacts with aqueous solutions or electrolytes.

b. **Electrochemical (wet) corrosion**

Wet corrosion causes the greatest amount of deterioration of materials. It requires the presence of water or other aqueous solutions (electrolyte) and a pathway for the transport of electrons [5].

b.1. Galvanic corrosion

This type of corrosion occurs due to the galvanic coupling of dissimilar metals such as metal restorations with different compositions, are placed in adjacent or opposing teeth. The less corrosion resistant metals become anode and corrode. When the two metals brought into contact, there is a sudden short-circuit formed that may cause what is called galvanic shock.

This galvanic current can occur also between restorations of similar alloys (as an old amalgam filling and a freshly placed amalgam restoration), because they will never have the same surface composition or structure. The old amalgam restoration will act as a cathode, and the freshly placed amalgam restoration will act as anode and corrode. This is because the old amalgam restoration has already undergone corrosion and oxidation and reached passivity (build up of oxide layer) which hinders corrosion progression. The high stability of the oxide film formed on the previously corroded old amalgam restoration serves as a further corrosion inhibitor [29].

Galvanic corrosion also occurs within the same filling material because the chlorine ion concentration in the interior surface of a dental restoration is seven times higher than that of saliva, so the inner surface of the restoration will have a higher electrochemical potential [2, 5]. The electrochemical cell is generated due to the difference in the electrode potential created by the two metals and the presence of electrolytes, as saliva and tissue fluids (the dentinal fluid, soft tissue fluid, and blood). Galvanic corrosion can occur in heterogeneous dental alloys' surfaces, having differences in electrode potential such as eutectic alloys, cored structure, impurities at grain boundaries, and solder joints [2, 5].

N.B: For galvanic corrosion to proceed, the production of electrons at anode by oxidation reactions must be exactly balanced by consumption of electrons at cathode. This is a very important in determining the rate of corrosion process. If the consumption of electrons at cathode is higher than production of electrons at anode the higher will be the corrosion rate and vice versa.

- High anode/cathode ratio (favourable) → decreased corrosion rate.

- Low anode/cathode ratio (unfavourable) → increased corrosion rate.

b.2. Crevice corrosion

This type of corrosion occurs when there are variations in the electrolyte composition within the system. Crevice corrosion occurs at the interproximal areas due to food debris and bacteria that alter the electrolyte composition, to be different from normal saliva. Also, Crevice corrosion results from differences in the oxygen tension. Irregularities such as pits, scratches and cracks in restoration are covered by food debris and mucin so the bottom of the defect becomes poor in oxygen concentration and acts as anode [2]. This is because the decrease in the oxygen concentration or hypoxia stimulates and increases the metal degradation or ionization. Also, the low oxygen tension causes reduction in the pH, which in consequence will increase the corrosion speed [31].

Crevices are unavoidable in surgical implants because implants never have simple shapes and are never exposed to a homogeneous environment. The local fluctuation of ions is significantly increased at crevice sites in the screw or plate. In the small space the liquid and oxygen exchanges are very limited and thus, the surface undergoes active corrosion and cause further deterioration of the implant material [2, 5].

b.3. Pitting corrosion

Pitting is a form of extremely localized attack that results in holes on the surface. Pitting corrosion is a type of corrosion that is similar to crevice corrosion, but it can begin on a smooth surface, as a consequence to localized breakdown of the passive layer that cause pitting. This type of corrosion can be initiated due to occlusal wear, acidic environment, or aggressive chemicals such as chloride [2, 6]. The rate of crevice and pitting corrosion is very rapid, because the area of the anode region is significantly smaller than that of the cathode. This unfavourable anode/cathode ratio causes rapid corrosion known as localized corrosion. Consequently, failure occurs much more rapidly than that expected from a uniform surface attack [2].

N.B: Uniform corrosion is defined as "a chemical or electrochemical reaction that proceeds uniformly over the entire exposed surface or over a large area".

b.4. Intergranular corrosion

Also termed intercrystalline or interdendritic corrosion. As the grain boundaries are known with its highly reactive nature, intergranular corrosion occurs adjacent to grain boundaries with little corrosion within the grains. This type of corrosion occurs due to the inhomogeneity. This type of corrosion is common in stainless steels.

It has been confirmed that prolonged heating of 18/8 stainless steel between 400 and 900 ◦C, during heat treatment or Welding, subjects the alloy to intergranular corrosion. This is due to the formation of small precipitate of chromium carbide along the grain boundaries. As a result, a chromium-depleted zone is formed adjacent to the grain boundary, which is known as sensitization/weld decay. So, the grain boundary region is now highly susceptible to corrosion [1, 7].

b.5. Selective leaching

Selective leaching is the removal of one element (the less noble component) from a solid solution alloy by galvanic corrosion process. Examples: leaching of Zinc in brass alloys (dezincification) in which zinc is selectively leached from a copper–zinc brass alloy. This process produces a weakened porous copper structure. Copper or Silver in Gold alloys, and Molybdenum in Nickel- Molybdenum alloys. Elements in an alloy that are more resistant to the environment remain behind.

There are two mechanisms for selective leaching: (1) Two metals in an alloy are dissolved; then one metal redeposits on the surface of the surviving elements. (2) One metal is selectively dissolved, leaving the other metals porous behind. The first system is involved in the dezincification of brasses, and the second system is involved when molybdenum is removed from nickel alloys [1, 2, 5].

b.6. Erosion-corrosion

Erosion corrosion is the acceleration or increase in rate of deterioration of a material due to the combined action of chemical attack and mechanical abrasion or wear. This type of corrosion arises because of the relative movement between a corrosive fluid and the material surface. It includes cavitation damage and fretting corrosion. Cavitation damage is not important in dentistry [5].

Fretting corrosion occurs in metal alloys that form a passive layer. The abrasive or wear action may erode the film, leaving an exposed metal surface. Erosion-corrosion is common in dental titanium implants [12]. During insertion of an implant, some degree of abrasion between fixing screws and implants or between tools and implants occur. Another source of abrasion on implant structures, is functional loading, which induces relative movement between the material (screw hole) and the screw head (i.e., the rubbing movement, disrupts the protective surface films). This action stimulates the chemical activities at the surface, leading to oxidation, active corrosion, or re-passivation [13].

b.7. Stress corrosion

Stress corrosion cracking is "the cracking caused by the simultaneous presence of tensile stress and a specific corrosive medium". Stress can accelerate the corrosion process because the internal energy is increased. Stress can be externally applied as restorations are subjected to heavy compressive, shear and bending forces during mastication. Also, it can be a residual stress as in cold worked structures due to permeant deformation, burnished restorations, or atoms at grain boundaries. Thus, an electrolytic cell is formed between the stressed and unstressed metal portions. For most metallic dental appliances, stress corrosion occurs mostly during fatigue or cyclic loading. Small surface irregularities act as sites of stress concentration (stress raisers), so the effects of fatigue and corrosion are combined [2, 5].

b.8. Hydrogen embrittlement

Also known as hydrogen assisted cracking or hydrogen induced cracking. It is the embrittlement of a metal by hydrogen. The atomic hydrogen is harmful to the toughness of iron and steel. It is a low temperature effect (near ambient temperature) because most metals are immune to hydrogen embrittlement above approximately 150°C. For hydrogen embrittlement to occur, a combination of three conditions are required: (1) the presence and diffusion of hydrogen atoms or ions, (2) a susceptible material and (3) stress 24.

Various metal alloys, as steels, experience a significant reduction in ductility and tensile strength when atomic hydrogen penetrates into the material, and diffuses interstitially through the crystal lattice. When these hydrogen atoms recombine in the voids of the metal matrix to form hydrogen molecules, they create internal pressure leading to crack formation (hydrogen induced cracking) [1, 15].

Copper alloys which contain oxygen can be embrittled if exposed to hydrogen. The hydrogen diffuses through the copper and reacts with inclusions of Cu_2O, forming H_2O, which then forms bubbles at the grain boundaries. This process can cause the grains to be forced away from each other causing embrittlement. Also alloys of vanadium, nickel, and titanium can absorb significant amounts of hydrogen. This can lead to large volume expansion and damage to the crystal structure leading to the alloys becoming very brittle [16].

N.B: Heating metals above 150°C causes the hydrogen within the metal to diffuse out through heat treatment, that's why hydrogen embrittlement is a low temperature effect.

b.9. Biocorrosion (Microbiologically influenced corrosion)

It is "the deterioration of the metal surface due to the direct or indirect activity of living microorganisms" [17]. This biological corrosion is influenced by the nature of their surrounding environment. An aggressive or electrochemically active environment can greatly increase the rate of corrosion of a metal. Biological corrosion is not considered actually a type of corrosion. But the activity of living organisms (bacteria, algae) can increase the corrosion process by affecting the anodic and cathodic reactions and the protective passive layer. It can also create corrosive conditions by producing deposits. Two types of microorganisms can cause this type of corrosion.

These are aerobic (oxygen using) and anaerobic (nonoxygen using). Anaerobic organisms are most responsible for corrosion of iron and steel [5]. Sulphate-reducing bacteria are the most important anaerobic bacteria that influence the corrosion of steels, Co-Cr-Mo and Ti- 6Al-4V alloys. These bacteria reduce inorganic sulphates to sulphides in the presence of hydrogen, where hydrogen is provided by the cathodic reaction or present in the general environment.

The reaction is according to the following equation:

$$SO_4^{-2} + 8H \longrightarrow S^{-2} + 4H_2O$$

Iron then reacts with sulphides according to the following reaction:

$$Fe^{+2} + S^{-2} \longrightarrow 2FeS$$

The presence of iron sulphide in rust is often an indication that corrosion was influenced by sulphate-reducing bacteria [17, 18, 19].

Factors affecting corrosion resistance of metals

Wide variety of factors influence the corrosion behavior of metals, including both the material and the surroundings. Changes in these variables can have an effect on the mode and rate of metal ion release. These factors include:

1. Presence of chloride and other ions

The presence of chloride affects the corrosion of metallic restorations and implants. Chloride ions can easily penetrate the passive film due to their high diffusivity. Leading to its accumulation at the metal/film interface. Chloride ions could further react with the metal substrate at the metal/film interface to form metal chloride. Rupture of the surface film due to volumetric expansion, because the molar volume of metal chloride is larger than that of the metal oxide in the surface film. Thus, pit nucleation occurs.

During the pitting process, the pH value of the electrolyte in the pit tends to decrease due to the formation of H+ ions. Once the pH value within the pits drops below a critical value, the repassivation behaviour of passive film is inhibited, and pits can further develop into stable pitting.

Fluoride ions are also very corrosive as chloride ion, and the corrosion ability of these ions is again enhanced when both are present together in the corrosive medium. Fluoride ions are very aggressive on the protective TiO 2 film formed on Ti and Ti alloys. When a Ti restoration is used, fluoride gels should be avoided because it creates an acidic environment that leads to the degradation of the titanium oxide layer and possibly inhibits osseointegration [20, 21].

2. Surface Topography

Most failures due to corrosion are known to initiate at the surface. The more homogenous is the surface, the higher is the corrosion resistance. Surface roughness offers a decrease in metal resistance to corrosion and increase the number of pits on the surface which act as nucleating sites for corrosion [20].

3. Dissolved oxygen, pH & bacteria

The corrosion behavior of metals is also influenced by oxygen content, pH, and presence of microorganisms. The effect of Oxygen Tension: The deposition of the biofilm leads to reduction in the amount of oxygen reaching the metal surface beneath the biofilm compared to the other areas. Areas of low oxygen tension act as anode, which undergoes corrosion releasing metal ions into the saliva.

These metal ions combine with the end-products of the bacteria, along with chloride ion in the electrolyte (saliva) to form more corrosive products like MnCl2, FeCl2 favoring further corrosion [22]. The PH influences the metal release in different ways, either directly through dissolution or indirectly by changing the extent of adsorbed complex agents or proteins. Generally, the release of metallic ions increases as the pH of the solution decreases. In case of base metal alloys [22]. Bacteria have great effect on the corrosion resistance of dental metallic material. The corrosion behavior of metals in the presence of Streptococcus mutans and its byproducts was found to be increased. This could be explained by the attachment of microbes on implants disturbs the passivity of passive metal. Furthermore, the formation of organic acids by bacteria lead to reduction in pH leading to a favorable environment for corrosion [22].

Factors Affecting rate of corrosion

1. Electrode Potential of individual metal. The more the reactivity of the metal, the higher will be the corrosion.
2. Relative position of anode to cathode in the Electromotive force of series: increasing the difference in electrode potential between anode and cathode will increase the rate of corrosion.
3. Increasing the anode/cathode ratio will decrease the rate of corrosion process as consuming the electrons at the cathode less than producing of electrons from anode
4. Type of electrolyte: normal saliva is weak electrolyte so the rate of corrosion will be lower when compared to rate of corrosion in acidic electrolyte.
5. Concentration of positive ions in the electrolyte. By increasing the ions in the electrolyte, the rate of corrosion is increased.
6. A rise in temperature (within a reasonable limit) increases the rate of corrosion [23].

Adverse effects of Corrosion Products

Most cast dental restorations are made from alloys or commercially pure titanium. It has been documented that metallic dental restorations release metal ions mainly due to corrosion. Those metallic components may be locally and systemically distributed and could play a role in the etiology of oral and systemic pathological conditions. The quality and quantity of the released cations depend upon the type of alloy and various corrosion parameters [24].

1. Allergic Reactions

Local and systemic allergic reactions to many metals have been observed. Of all the metals the most allergens are Ni, Cr, Co and Hg. While as Aluminium and Titanium are said to be rare allergens. These allergens usually cause allergic contact dermatitis which appears as swelling, redness, burning sensation, ulceration, and sometimes reaches necrosis [25].

2. Metallic Taste

The symptom of metallic taste has been reported and related to the release of ions from the metal. Patients with bruxism are likely to be more susceptible to metallic taste [25].

3. Discoloration of teeth

It has occurred mainly with amalgam fillings and metallic screw posts. With amalgams, Grey pigmentation is common in teeth with amalgam restorations. Corrosive amalgam discolors the dentine by formation of silver sulfide. While with the screw posts, copper and zinc were detected in both the dentin and enamel and the surrounding soft connective tissue. It was found also that Mercury, lead, and silver nitrate salts used in dentistry can cause a grey staining in teeth [26, 27].

4. Galvanic pain

It has been reported from the contact of dissimilar-alloy restorations. In this case, instantaneous current flows through the external circuit causing pain [28].

5. Oral lesions

Permanent irritation to the oral mucosa can be generated by the shape and location of the prosthesis in the mouth. Corrosion of a metallic prosthesis can change the smooth surface of the prosthesis to a rough & sharp surface which increases the irritation in the opposing tissues [11].

6. Bone Loss

Ti alloys contain significant amounts of alloying elements that affect osseointegration mainly due to corrosion products containing aluminium and vanadium. It was found that macrophages were present in the peri-implant tissue with the metallic ions. Thus, corrosion process plays an important role in implant failure. Moreover, presence of free titanium ions in the peri-implant tissue inhibits growth of hydroxyapatite crystals. These processes lead to local osteolysis and loss of clinical stability of the implant [21].

7. Metal Fracture

Corrosion can severely limit the fatigue life and ultimate strength of the material leading to mechanical failure and metal fracture [21].

Protection Against Corrosion

1. Increase noble metal content

As noble metals have higher electromotive potential when compared to any other metals used in the oral environment, it can be added to alloys to reduce corrosion. At least half of the atoms in the alloy should be noble metals (gold, platinum, and palladium) to ensure the resistance against corrosion. Palladium is found to be effective in reduction of silver sulfide formation for alloys containing silver [7].

2. In Base metal alloys

Passive metals develop a thin, adherent, highly protective film when they react with the environment. Chromium, titanium and aluminium are examples of such metals. Base metal alloys containing chromium are resistant to corrosion because of the passivity of chromium (not less than 20%) to form adherent oxide layer [7].

3. Polishing of restorations

Polished smooth metallic restorations like amalgam and cast metal minimizes corrosion [7].

4. In presence of two dissimilar metals

Increase anode/cathode ratio to decrease rate of corrosion process, as consumption of electrons at cathode is less than production of electrons at anode. Paint cathode surface by varnish coating [7].

5. Maintenance of oral hygiene

Conclusion

Materials used in the oral cavity are constantly exposed to fluctuating physical and chemical environment that includes temperature and components like saliva, salts, foods, acidic and alkaline liquids, and drugs. Plaques and food adhere to teeth and filling material which means several forms of bacteria and their products are always present inside mouth. Also, fluoride ions interfere in the titanium oxide formation which causes changes in the passive layer formation. This makes the oral environment a highly conductive medium for tarnish & corrosion.

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