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Corrosion of galvanic pairs of Co-Cr alloys with high-copper silver amalgam using Mansfeld formulas

Corrosión de pares galvánicos de aleaciones Co-Cr con amalgama de plata de alto cobre usando las fórmulas de Mansfeld

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ABSTRACT

In the restoration of lost or damaged dental pieces, different alloys are frequently used, leaving the mouth exposed to electrical currents that circulate through saliva and dental fluids. In the present work, electrochemical methods are used to determine the corrosion rate of galvanic pairs of a high copper silver amalgam with Co-Cr dental alloys in artificial saliva. It is observed that when the difference in corrosion potentials of dissimilar alloys are small (less than 100 mV_{ecs}), Evans diagrams do not give good results, so the Mansfeld correction formulas must be used. Thus, it is found that the most resistant to corrosion is the pair formed between the amalgam and the bulk Co-Cr alloy ($PG_{1,3}$) and that the corrosion products released in greater quantity to the electrolyte are those that contain ions of Co, Cr, Ag, Sr and Cu.

Keywords: Corrosion, dental alloy, polarization curve, artificial saliva, galvanic pair.

RESUMEN

En la restauración de piezas dentales perdidas o deterioradas se usan frecuentemente diferentes aleaciones quedando la boca expuesta a corrientes eléctricas que circulan a través de la saliva y los fluidos dentarios. En el presente trabajo se utilizan métodos electroquímicos para determinar la velocidad de corrosión de pares galvánicos de una amalgama de plata de alto cobre con aleaciones dentales Co-Cr en saliva artificial. Se observa que cuando la diferencia de potenciales de corrosión de las aleaciones disímiles son pequeñas (menos de 100 mV_{ecs}), los diagramas de Evans no dan buenos resultados, por lo que deben usarse las fórmulas de corrección de Mansfeld. De éste modo se encuentra que el más resistente a la corrosión es el par formado entre la amalgama y la aleación Co-Cr a granel ($PG_{1,3}$) y que los productos de corrosión liberados en mayor cantidad al electrolito son los que contienen iones de Co, Cr, Ag, Sr y Cu.

Palabras clave: Corrosión, aleación dental, curva de polarización, saliva artificial, par galvánico.

1. INTRODUCTION

Dental alloys for melting and casting are available in the market in a wide variety of compositions and mechanical properties that depend on their subsequent application (crowns, bridges, inlays, etc.). The most traditional are the noble alloys that contain not less than 75% of gold and metals of the platinum group. These alloys do not suffer deterioration of their properties over time or lose their aesthetic appearance. However, they have high density, low elastic modulus and are excessively expensive [1-4].

To replace alloys with a high gold content, non-precious alloys have been developed, which have been used since the 40s of the last century in developed countries such as the United States and Germany, among these alloys are those of Ni-Cr, Co-Cr, stainless steel and lately the titanium base. Co-Cr alloys have high

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resistance to heat, corrosion, wear, discoloration and are non-magnetic [5,6].

Co-Cr alloys are frequently used for fixed and partially removable denture frames [7-9]. In the manufacture of some complex frames and repair shops, apart from the metal segments of the frames these have to be joined using solder whose melting point is lower than the pieces to be welded [10,12]. On the other hand, in the oral cavity of many people there are dental pieces filled with amalgams, others have crowns, bridges, incrustations, implanted structures, or in the form of wires to correct masticatory defects by means of orthodontic appliances [1-3]. The use of different metal biomaterials in the mouth of the users gives rise to oral galvanism, a phenomenon that has been studied in different electrolytic media that simulate natural saliva[13-21].

The potentials and current densities of corrosion of the galvanic pairs are determined using Evans diagrams. However, this method does not always give good results, obtaining in some cases corrosion current densities, of the galvanic couplins under study, values lower than the current densities of the non-coupled alloys. For this reason, in this work we have studied the corrosion of galvanic pairs of Co-Cr alloys with high copper silver amalgam in artificial saliva using the Evans method, the Mansfeld correction formulas and we have analyzed by EDAX the corrosion products in the cavity oral simulation.

2. MATERIALS AND METHODS

2.1 Materials

A high copper silver amalgam and three Co-Cr dental alloys whose chemical composition and tradename is shown in Table 1 have been used [22-24].

Table 1: Chemical composition of high copper silver amalgam and dental alloys Co-Cr in weight %.

MATERIAL	TRADENAME	CHEMICAL COMPOSITION (WEIGHT %)
Dental Mercury	Mercurio K - Dent	Hg 99.99
Amalgam of high Cu	Duralloy S	Ag 45.0 - Sn 31.0 - Cu 24.0
Alloys Co - Cr	Vera PDN	Co 63.5 - Cr 27.0 - Mo 5.5 - Fe 2.0 – Ni 0.99 – Si >1.0 – Mn >1.0
	Co - Cr (bulk)	Co 65.4 - Cr 26.0 - Si 4.5 - Fe 1.4 - Ni 0.8
	Premium	Co 59.9 - Cr 24.0 - W 8.5- Nb 2.2- V 2.2- Mo 0.95- Fe 1.5 – Mn 0.6 – Si >0.5

2.2 Preparation of test pieces

The Co - Cr test specimens were prepared by the method of the lost wax with an oxygen - butane - propane flame and subjected to a centrifugation process [25]; these were sheets of 1 cm^2 of surface per 0.2 cm of thickness. The silver amalgam specimens were prepared according to the manufacturers instructions in the form of cylinders 0.5 cm high by 0.8 cm in diameter.

2.3 Electrolyte

The electrolyte used in electrochemical tests is an experimental saliva that reproduces the electrochemical behavior of natural saliva [26], whose formula is shown in Table 2. The electrolyte was prepared with deionized water of 18.2 $\Omega \cdot$cm electrical resistivity and with reagents of analytical grade. The pH of the solution is 6.5; to prevent this value from being modified, $KHCO_3$ must be added shortly before starting the tests.

Table 2: Chemical composition of experimental saliva. Concentration in (g/L) [26].

NaCl (g/L)	KCl (g/L)	$CaCl_2.2H_2O$ (g/L)	KH_2PO_4 (g/L)	$Na_2HPO_4.12H_2O$ (g/L)	KSCN (g/L)	$KHCO_3$ (g/L)	$H_8C_6O_7$ (g/L)
0.600	0.720	0.220	0.680	0.856	0.060	1.500	0.030

2.4 Electrochemical methods

To determine the corrosion current density of the galvanic pairs of the Duralloy amalgam with the Co-Cr alloys, the open circuit corrosion potentials of the alloys under study were measured and then the polarization curves of these materials were drawn following way:

- **Potential Measures** - The electrode potential of each alloy was measured against a saturated calomel electrode in a three-electrode cell with a *Princeton Applied Research model 173* potentiostat in aerated artificial saliva at 25 °C. Measurements were done five times with different specimens of each alloy, previously stabilizing the potential for one hour in the solution, aerating it continuously with a fish tank aerator at about 80 bubbles per minute.
- **Polarization curves -** The polarization curves in artificial saliva were plotted in triplicate using the same cell and the same potentiostat as the one used to measure the electrode potential. The reference electrode was also saturated calomel and the platinum counter electrode. The scanning speed was 12 mV/min controlled with a Universal Programmer PAR 175 and the recording of the curves was performed with an XT PAR model REO 151. Before the beginning of the curves the corrosion potential was measured in the same way than in the previous section. The anodic and cathodic curves of each alloy were plotted separately with different probes starting from the electrode potential.
- **Potential corrosion of galvanic pairs** - Dental alloys corrode in the oral environment by the action of saliva and oral fluids, so that when they are electrically coupled they are polarized and corrode at a new speed. When metals A and B are coupled, the mixed potential of the galvanic coupling, $V_{corr.PG,}$ is at the intersection of the polarization curves where the total oxidation rate is equal to the total reduction velocity and the current density polarization is $I_{corr.PG}$ [27]; this process that allows to determine the potential and the current density of the cupla is called the Evans method.

2.5 Microanalysis of corrosion products

Corrosion products were analyzed by the Energy Dispersive Spectroscopy X-Ray (EDAX) Methods, which consists of analyzing the characteristic X-rays emitted by a sample reached by a beam of high-energy electrons from a scanning electron microscope, allowing the identification of the elements composing said sample through a multi-channel analyzer. In this work, a scanning electron microscope Philips SEM 500, from the Electronic Microscopy Laboratory of the Materials Department of CNEA, to which an EDAX DX4 dispersive detector in X-ray energy is attached, and whose limit of detection is approximately one micron cubic.

3. RESULTS AND DISCUSSION

3.1 Potential Corrosion

Table 3 shows the open-circuit corrosion potentials of the alloys under study. The alloys of Co - Cr are ordered according to their corrosion potential, forming an electrochemical series in aerated artificial saliva. The average standard deviation σ(Vecs) calculated according to standard G: 16-95 ASTM [28] is also shown.

The most active Co-Cr dental alloy is Premium whose corrosion potential is -0.259 V_{sce} probably due to the lower Cr content (24%). This alloy will always be an anode when it forms galvanic pairs with any of the dental alloys considered in this work. The amalgam of high copper (Duralloy) occupies an intermediate place, in such a way that when forming galvanic pairs with the alloys under study, it will act sometimes as an anode and sometimes as a cathode.

Table 3: Open circuit corrosion potentials of high copper silver amalgams, Co-Cr dental alloys in aerated artificial saliva and standard deviations (σ). The code numbered from 1 to 4 is to accurately identify the galvanic pairs.

MATERIAL	TRADENAME	CODE	V_{corr} (V_{sce})	σ (V_{sce})
Amalgam of high Cu	Duralloy S	1	-0,230	0,012
Alloys Co-Cr	Vera PDN	2	-0,216	0,007
	Co-Cr (bulk)	3	-0,245	0,007
	Premium	4	-0,259	0,013

3.2 Polarization curves

Polarization curves were determined under static conditions at room temperature (25 °C). Figure 1 shows the anodic and cathodic polarization curves of the alloys considered in this work. The high copper amalgam (Duralloy S) has a passive zone that extends from 0.260 V_{sce} to 1.100 V_{sce} with an average current density of 3.80 µA / cm^2; from 1.100 V_{sce} the current density increases rapidly due to the release of oxygen by the decomposition of the solution.

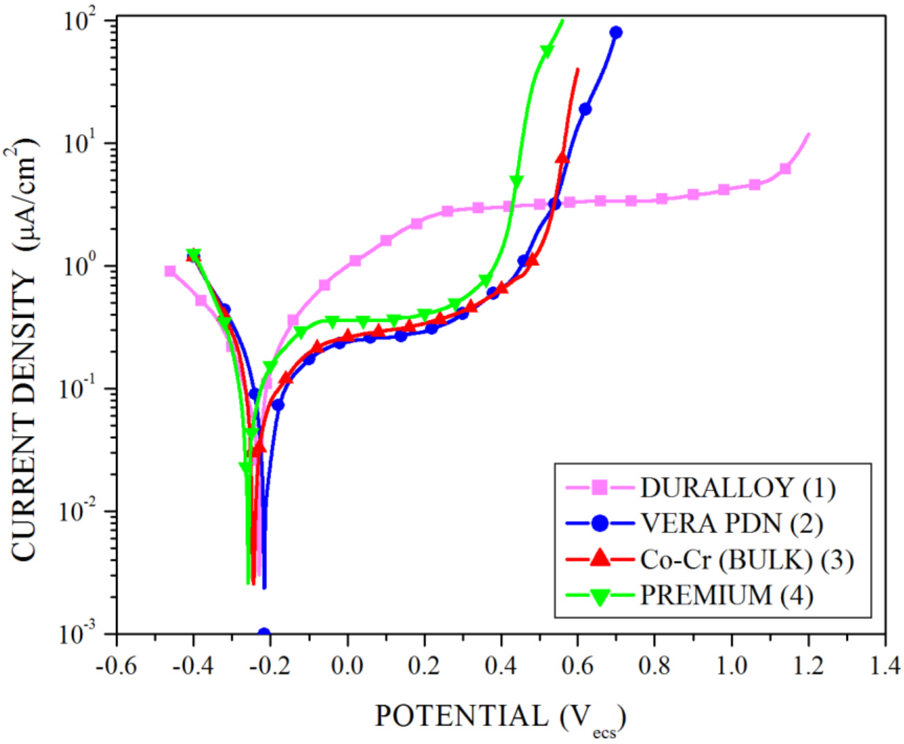

Figure 1: Polarization curves of Co-Cr alloys in aerated artificial saliva and Duralloy amalgam.

In Figure 1 it is also observed that the passive zone of the Co-Cr alloys extend approximately from -0.080 V_{ecs} to their rupture potentials of 0.300 V_{ecs}, with average current densities of 0.25 µA / cm^2, 0.30 µA / cm^2 and 0.34 µA / cm^2 for Vera PDN, Co-Cr in bulk and Premium respectively. There is clearly a decrease in the nobility of the alloys due to the lower content of chromium and molybdenum. In general, the corrosion resistance of Co-Cr alloys is due to the presence of Cr (III) oxyhydroxide present in the passive layers of these alloys [29]. For all these alloys, the current density rapidly increases to higher potentials than those of the passive zone, due to the dissolution of the poor interdendritic zone in Cr.

3.3 Potentials and densities of corrosion current of galvanic pairs of dental alloys

The potentials and instantaneous corrosion current densities of the galvanic couplings between different materials are obtained using the Evans method described above. In this case they are superimposed: (1) the cathodic curve of the high copper silver amalgam (Duralloy) with the anodic curves of the Premium and Co-Cr bulk alloys and (2) the anodic curve of the high copper amalgam (Duralloy) with the cathodic curve of Vera PDN, as shown in Fig. 2

Table 4: Corrosion current density of isolated dental alloys, potentials and corrosion current densities of Duralloy galvanic couplings with dental alloys Co-Cr by the Evans method. The (*) indicates that the Co-Cr alloy of the corresponding galvanic couple is corroded.

ALLOYS	I_{corr} (μA/cm^2)	Galvanic pair of high copper silver amalgam (Duralloy S)		
		CODE	$V_{corr.PG}$ (V_{sce})	$I_{corr.PG}$ (μA/cm^2)
Ag Amalgam:				
• Duralloy S	0.033			
Alloys Co-Cr:				
• Vera PDN	0.110	$PG_{1,2}$	-0.225	0.029
• Co-Cr (bulk)	0.128	$PG_{1,3}$	-0.236	0.019*
• Premium	0.113	$PG_{1,4}$	-0.240	0.055*

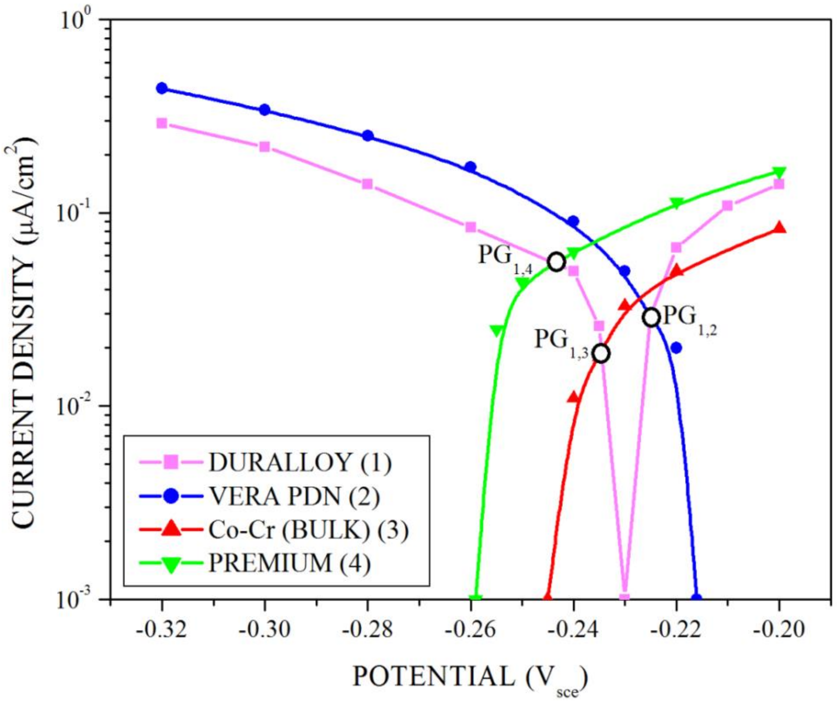

Figure 2: Superposition of Duralloy amalgam polarization curves with the anodic curves of the Premium and Co-Cr alloys in bulk, and the anodic curve of Vera PDN in aerated artificial saliva.

Table 4 also shows that the corrosion current densities of Duralloy's galvanic pairs with the Co-Cr alloys under study are lower than the current densities of the anode alloys: 1) Premium and Co- Cr in bulk, 2) Vera PDN. That is, it violates the condition of the galvanic pair theory that states that the current density of a galvanic pair is greater than the current density of the uncoupled anode. This would lead to the erroneous assumption that the galvanic coupling of these alloys increases the corrosion resistance of the galvanic couplings under study.

3.4 Correction of results of galvanic current densities by the Mansfeld method

In the decade of the 70s of the last century, Mansfeld [30-32], systematically studied the behaviour of the galvanic pairs and the applicability of the overlap of the Evans diagrams for the determination of the galvanic currents and concluded that three cases should be considered: 1) Tafel behavior, 2) diffusional control and 3) small polarization ranges. In this paper we will deal only with the last case.

For small polarization ranges, coupling metal A to metal B (nobler than A) produces only a small displacement of the corrosion potential of metal A. Hence, the potential of the galvanic pair of the two different metals coupled, is located very close to the corrosion potential of the uncoupled anode and there is no Tafel type behavior; and both the anodic and the cathodic reactions occur at a rate significantly higher than that of the uncoupled anode.

In this case the galvanic current (Icorr.PG) is not equal to the dissolution current of metal A. This last current is the sum of the cathodic currents on the anode ($I_{A,\ c}$) and on the cathode ($I_{B,}$ c):

$$I_{A,\ a} = I_{A,\ c} + I_{B,\ c} \tag{1}$$

while the galvanic current is equal to the cathodic current on the metal B:

$$I_{corr\cdot PG} = I_{B,\ c} = I_{A,\ a} - I_{A,\ c} \tag{2}$$

from where

$$I_{A,a} = I_{corr.PG} + I_{A,c} \tag{3}$$

The magnitude of the current density of the galvanic pair ($I_{corr.PG}$) is determined with the Evans diagrams and the cathodic current density of the anode A is calculated with the formula:

$$I_{A,c} = I_{corr.A} \exp\left(- \frac{\left(V_{corr.PG} - V_{corr.A}\right)}{0{,}434\, b_c} \right) \tag{4}$$

Where $V_{corr.PG}$ is the corrosion potential of the galvanic pair, $V_{corr.A}$ and $I_{corr.A}$ are the corrosion potential and the corrosion current density of the decoupled anode; b_c is the cathodic Tafel slope of the anode material (A).

The polarization range ($V_{corr.PG}$ - $V_{corr.A}$) of the galvanic pairs of silver amalgams with non-precious dental alloys considered in this work is very small, less than 0.16 V_{sce}, so the formulas will be used (3) and (4) to determine the true dissolution current density of the anode ($I_{A,\ a}$) for the corrosion potential of the galvanic couple. We will illustrate the calculation of $I_{A,a}$, with an example.

For the $PG_{1,2}$ pair (Duralloy / Vera PDN), the galvanic current density (Table 4) is: $I_{corr.PG}$ (1,2) = 0.029 μA / cm^2 and the current density $I_{A,c}$ corresponding to the anode (amalgam Duralloy) is determined using the formula (4) and the data of tables 3 and 4:

$$I_{A,c} = (0.243\ \mu A/cm^2) \exp\left(-(-0.225+0.230)/(0.434 \times 0.167)\right) \tag{5}$$

from where

$$I_{A,c} = I_{1,c} = 0.227\ \mu A/cm^2 \tag{6}$$

The dissolution of the anode (Duralloy) to the corrosion potential of the $PG_{1,2}$ pair is obtained by formula (3):

$$I_{A,a} = I_{corr.PG} + I_{A,c} = 0.256\ \mu A/cm^2 \tag{7}$$

In this way, the actual values of the dissolution of the anode of the galvanic pairs considered in this work are determined, which are shown in Table 5.

Table 5 shows that: 1) the corrosion current density of the galvanic couple in which the high copper amalgam (Duralloy) acts as an anode, evaluated with the Mansfeld formulas is greater than the corrosion current density of the uncoupled amalgam (0.033 μA / cm^2) in accordance with the PG principle that states that the current density of the galvanic couple ($I_{corr.PG}$) must be greater than the current density ($I_{corr.A}$) of the decoupled anode [32]. 2) the galvanic pairs in which the Duralloy amalgam acts as cathode, dissolves at a higher speed than the Premium and Co-Cr bulk alloys decoupled according to the PG principle mentioned above.

Table 5: Cathodic Taffel slopes and corrosion current densities of the Duralloy amalgam galvanic pairs with dental alloys by the methods of Evans and Mansfeld. The values with an asterisk (*) means that the Co-Cr alloys are dissolved and not the amalgam.

ALLOYS	b_c (V_{ecs})	CODE	$I_{corr.PG}$($\mu A/cm^2$) (Evans)	$I_{A,a}$ ($\mu A/cm^2$) (Mansfeld)
Amalgam Duralloy S	0,167			
Alloys Co-Cr:				
• Vera PDN	0,170	$PG_{1,2}$	0,029	0,256
• Co-Cr (bulk)	0,168	$PG_{1,3}$	0,019*	0,159*
• Premium	0,147	$PG_{1,4}$	0,055*	0,231*

3.5 Corrosion Products

Figure 3 shows the scanning electron photomicrograph (SEM) of the surface of the Duralloy high copper amalgam and the EDAX of its corrosion products, showing high peaks of Hg, Ag, Sn and Cu. The amalgam phases are also observed that according to Yusif studies [32], the Cu-Sn phase is less resistant to corrosion the grains do not come into contact with each other producing only surface corrosion. On the other hand, the corrosion products of high copper amalgams contain two elements identified as Cu_2O of red color and $CuCl_2.3Cu(OH)_2$ of green color. Finally, these amalgams emit a greater amount of mercury vapor than conventional amalgams because they lack the γ_2 phase [33, 34].

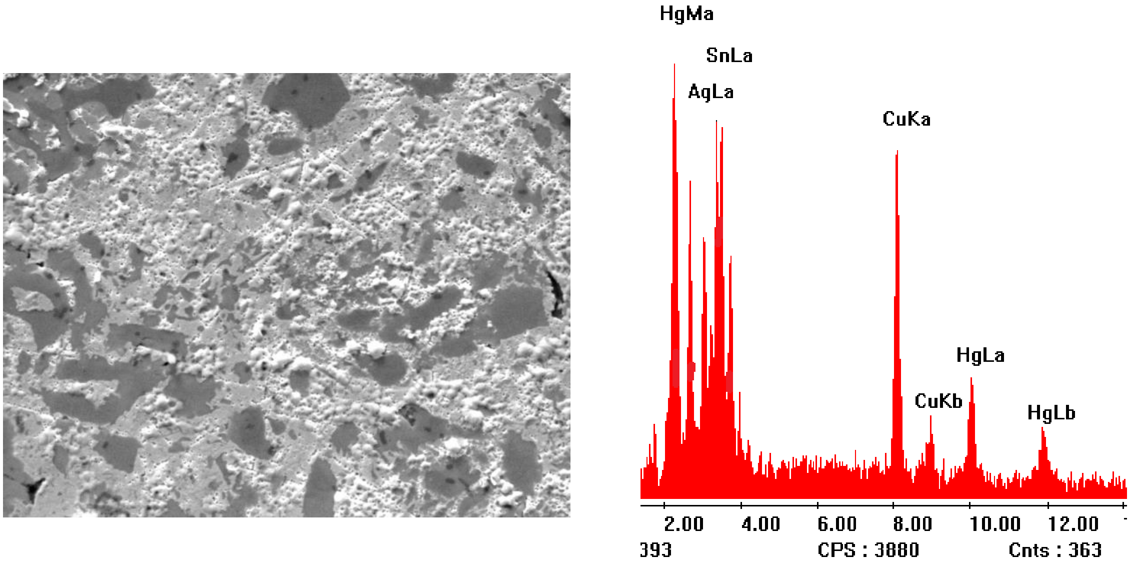

Figure 3: SEM micrograph and EDAX spectra of corrosion products on Duralloy (x1440).

Figure 4 shows the SEM and EDAX photomicrograph of the corrosion products of the Vera PDN alloy, observing large Co and Cr peaks and small Mn, Fe and Cu peaks. As oxygen and hydrogen are not determined by this method, it is assumed that oxides and hydroxides of these elements are formed in addition to chlorides and phosphates.

Figure 5 shows the SEM and EDAX photomicrograph of the bulk Co-Cr alloy corrosion products, observing large and small Co and Cr peaks, assuming that oxides and hydroxides of these elements are formed in addition to chlorides and phosphates.

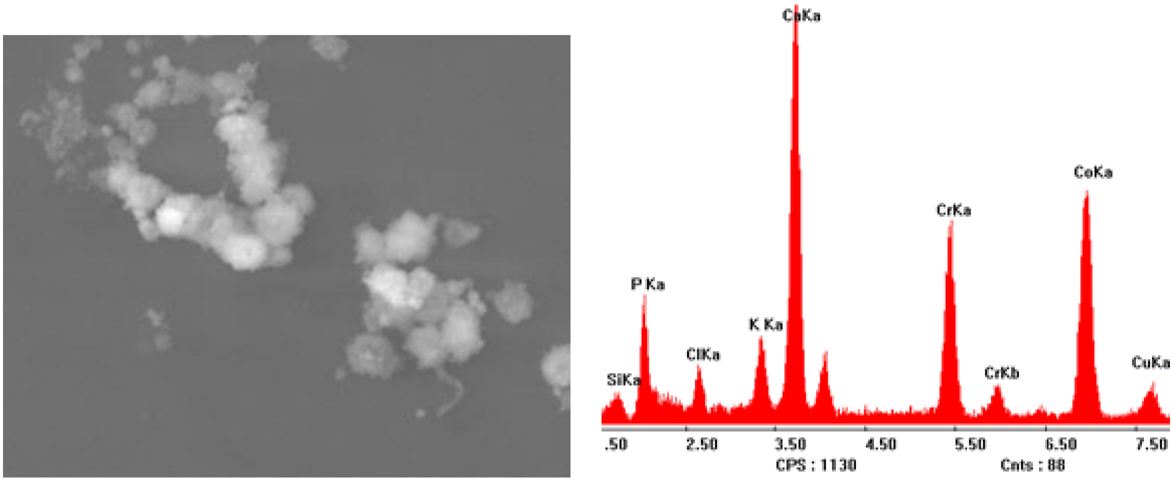

Figure 4: SEM micrograph and EDAX spectra of corrosion products on Vera PDN (x1440).

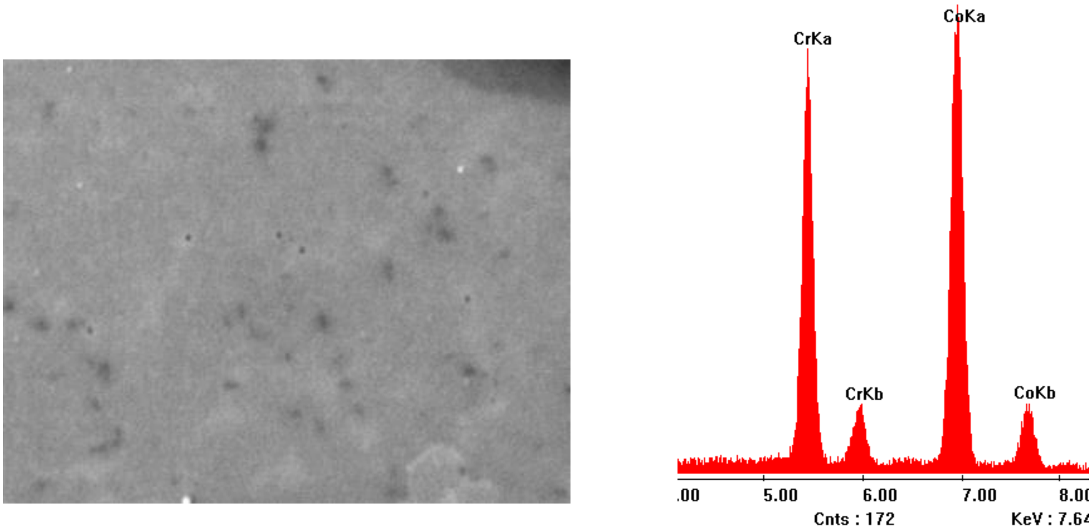

Figure 5: SEM micrograph and EDAX spectra of corrosion products on bulk Co-Cr (x1440).

Figure 6 shows the SEM and EDAX photomicrograph of the corrosion products of the Premium alloy, observing large and small Co and Cr peaks, so oxides and hydroxides of these elements must be formed in addition to chlorides and phosphates due to the components of the solution and presence of oxygen and hydrogen in the aerated electrolytic medium.

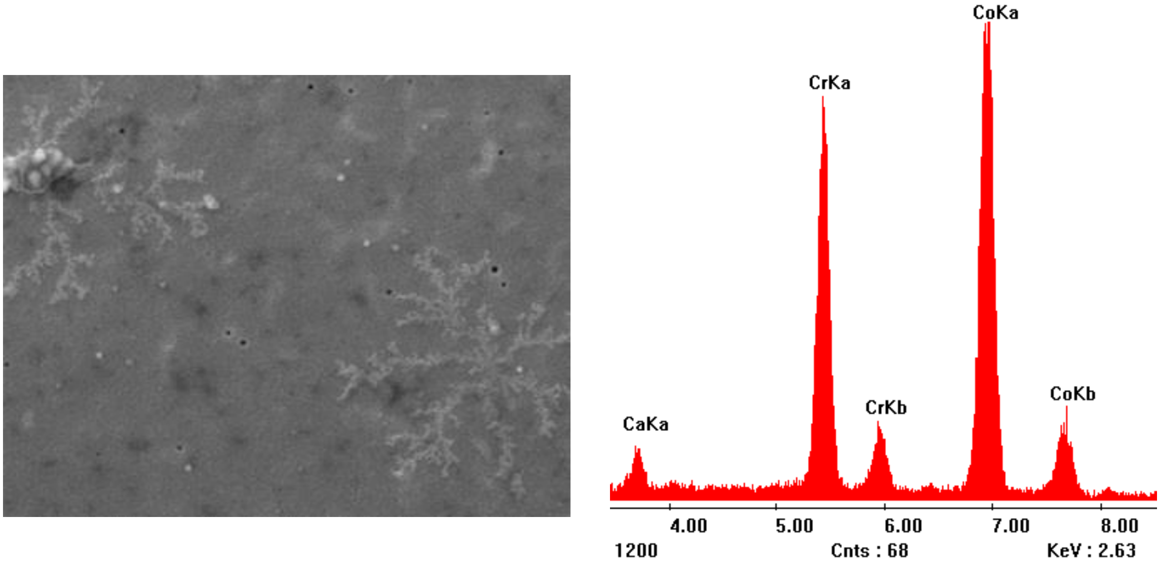

Figure 6: SEM micrograph and EDAX spectra of corrosion products on Premium (x1440).

No metal or alloy is completely inert in the living being. All eliminate ions from its surface in greater or lesser intensity. Co-Cr alloys produce chromium oxides but the only ion captured by the intracellular red blood cells is Cr^{6+} which then rapidly converts to Cr^{3+}[35].

Corrosion products that enter the body produce discoloration of adjacent soft tissues, allergic reactions and skin rashes in some people. Hansen in 2006 investigating the corrosion of Co-Cr-Mo alloy in Hank's solutions, showed that this alloy dissolves producing chromium and molybdenum oxides that affect the health of people [36,38]. On the other hand, Cu, Ag, Sn and Hg ions, released into the oral cavity by the corrosion of the alloys under study, affect the viability and proliferation of the lymphocytes. With time, the concentration of copper increases in the gums, producing inflammation, alteration of cellular immunity and oral homeostasis [37-40].

4. CONCLUSIONS

From the results obtained in this work we can conclude:

1. The amalgam of high copper silver (Duralloy) by forming galvanic pairs with the Co-Cr alloys will act as a cathode for having a nobler corrosion potential than these alloys in aerated artificial saliva, except with Vera PDN.
2. The experimental results show that for small polarizations the Evans method does not always give good results to predict the dissolution rate of the anodic component of the galvanic pairs, so it is necessary to use the Mansfeld correction formulas.
3. The corrosion products of the galvanic couplings under study release in greater quantity ions of Hg, Co, Cr, Ag, Sn and Cu which will pass from the mouth to the digestive tract and to different parts of the body through the circulatory system.

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