

COMPARATIVE EVALUATION OF THE CORROSION RESISTANCE OF SOME CO-CR ALLOYS USED IN METAL-CERAMIC TECHNOLOGY

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Abstract: Co-Cr alloys are still used in metal-ceramic technology on the large scale. The purpose of this study was to provide a comparative analysis of the corrosion resistance by the polarization resistance technique of two Co-Cr-based alloys (codified Co-Cr-P and Co-Cr-PW) produced by the same manufacturer, which have a minor variation in chemical composition. Corrosion tests were performed at a temperature of $37 \pm 0.5^\circ \text{C}$ in a bath with heating and recirculation, in artificial saliva (SA) Carter Brugirard at $\text{pH} = 7.58$. Both tested alloys showed an increased corrosion resistance when immersed in artificial saliva, proving a good corrosion resistance and therefore a good biocompatibility. The Co-Cr-P test showed superior electrochemical values and, implicitly, higher corrosion resistance.

Keywords: corrosion, dental alloys, artificial saliva.

Introduction

Although lately fixed prosthetic restorations made of all-ceramic materials [1-10] or of ceramics and zirconia [11-15]

have gained more and more ground, ceramic crowns and dental bridges deposited on metal skeletons of base alloys [16-18] continues to represent the "gold

standard" in fixed prosthetics at affordable costs for most of the world's population.

The metal-ceramic restorations from non-noble alloys through the metal framework substructure confer resistance to the structure, while the veneering ceramic offers a superior aesthetic aspect, which successfully fits into the dento-facial harmony.

Co-Cr-based dental alloys have gained ground over other alloys due to possible allergic responses after exposure to Ni or Be [19-24], because of the release of these ions into the oral environment by corrosion of dental alloys over time.

The purpose of this study was to provide a comparative analysis of the corrosion resistance of two Co-Cr-based alloys produced by the same manufacturer, which have a minor variation in chemical composition. The most important difference is that the first alloy contains Mo

(4 %wt) and the second one instead Mo contains 4 %wt Fe. The corrosion tests were carried out under temperature and pH conditions that mimic the oral environment.

Materials

Two cast commercially Co-Cr-based dental alloys manufactured by Kulzer, respectively Heraenium® P and Heraenium® PW were chosen to evaluate the corrosion resistance. These materials are commonly used for the metal-ceramic restorations.

The chemical composition of commercial alloys is given in Table 1. It can be clearly seen that there is a minor variation in chemical composition of the selected alloys. The Co-Cr-P alloy has a higher amount of Co, a lower amount of W as compared to Co-Cr-PW alloy. Also, Co-Cr-PW alloy does not contain Mo, but has a slight amount of Fe.

Table 1. Chemical composition of cast Co-Cr alloys according to the manufacturer (%wt)

No.	Trade name	Elements (%wt)							Codification
		Co	Cr	W	Si	Mn	Fe	Mo	
1.	Heraenium® P	59	25	10	1	0.8	-	4	CoCr-P
2.	Heraenium® PW	55	24	15	1	0.8	4	-	CoCr-PW

The chemical composition of the Co-Cr alloys used could be identified using an energy dispersive X-ray spectrometer (EDS) as part of the scanning electron microscope - SEM (Phenom ProX model,

manufacturer PhenomWorld, The Netherlands). In order to obtain the chemical composition, the analysis was performed on an area of 270 x 270 μm^2 and the obtained results are found in table 2.

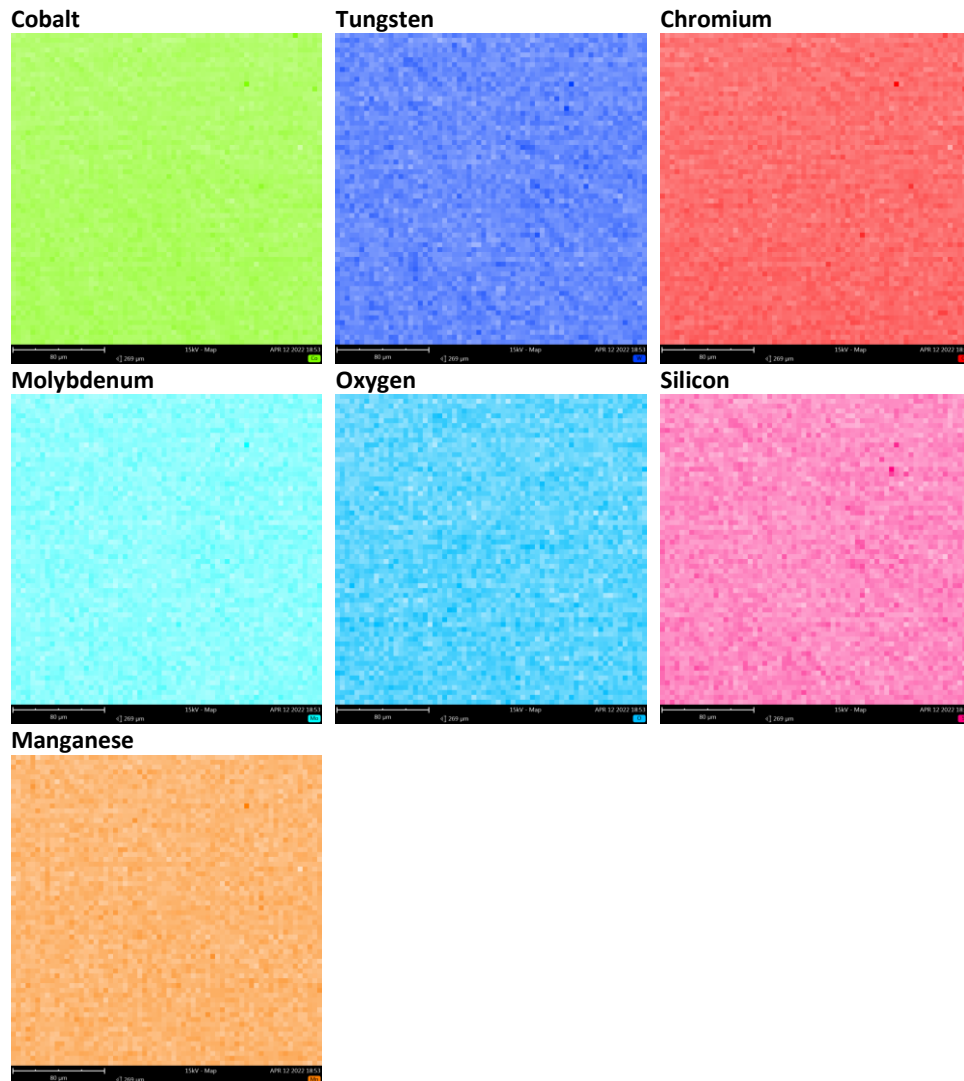
Table 2. The chemical composition of the Co-Cr alloys obtain by EDS

No.	Sample	Elements (%wt)							
		Co	Cr	W	Si	Mn	Fe	Mo	O
1	CoCr-PW	53.77	23.42	15.07	1.9	0.51	2.23	-	3.1
2	CoCr-P	57.63	23.58	9.7	2.2	0.48	-	3.99	2.42

From the analysis of the chemical compositions obtained by EDS it can be said that the investigated samples are from the class of Co-Cr alloys having values close to those specified by the manufacturers. The oxygen in the chemical composition obtained by EDS comes from

the oxide formed on the surface of the alloy and which protects it against corrosive agents.

Figures 1 and 2 show the EDS spectra obtained and the distribution of the elements on the investigated surface for the two Co-Cr alloys.



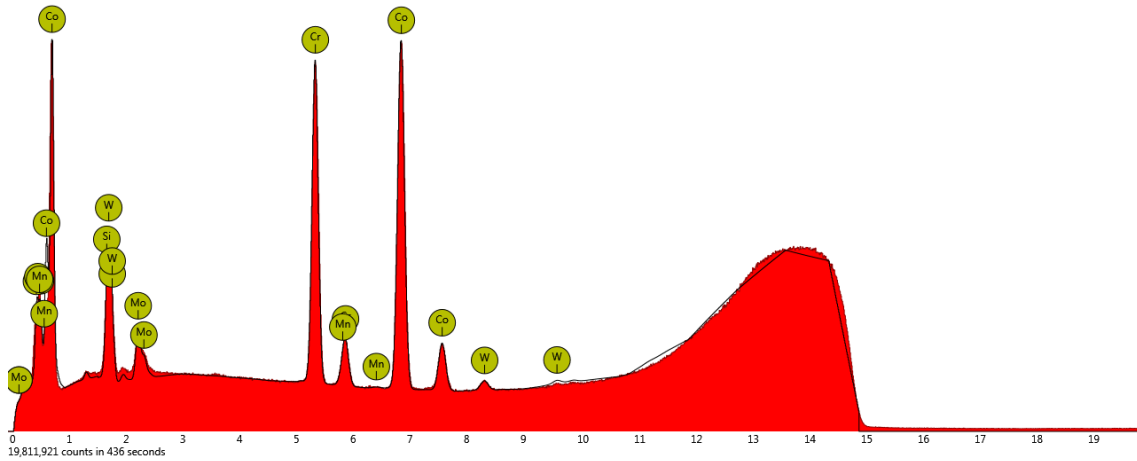
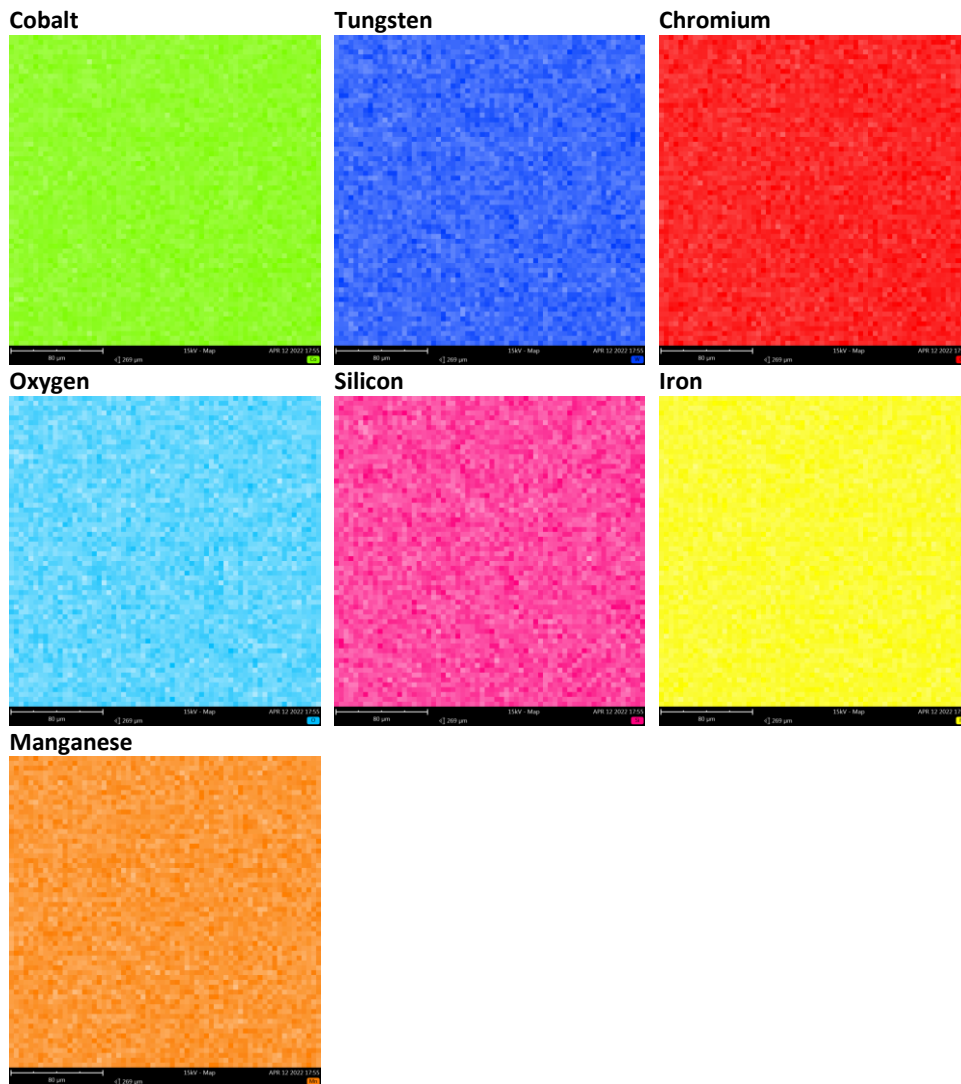


Fig. 1. EDS spectrum and distribution of elements for the CrCr-P sample



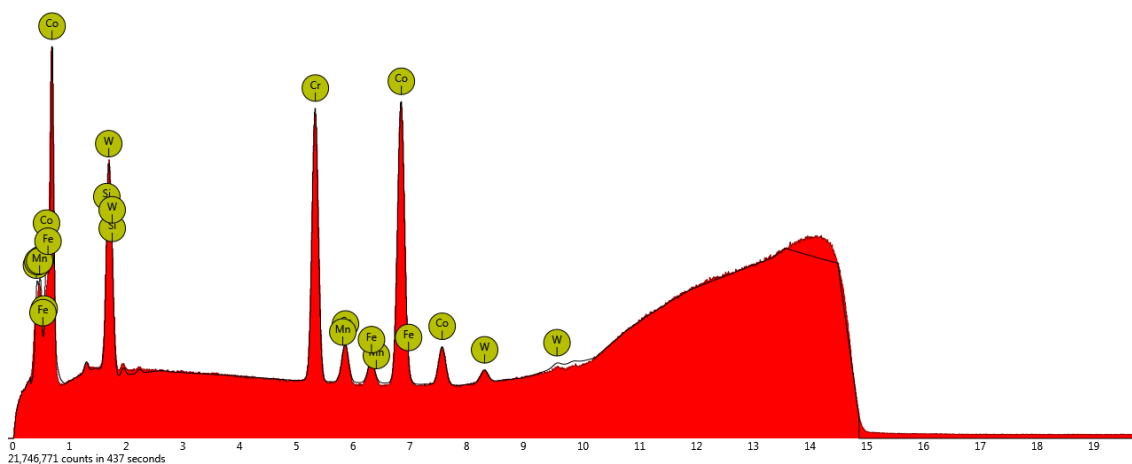


Fig. 2. EDS spectrum and distribution of elements for the CrCr-PW sample

Preparation of the samples

The cylindrical samples with 8 mm diameter were cut with a abrasive disk from commercially available blocks (manufactured by casting). An isolated copper conductor was glued to the cylindrical samples and individually the samples were embedded in epoxy resin. The samples are having an exposed area of 5 mm².

Potentiodynamic polarization tests

Corrosion behavior testing was performed with a Potentiostat / Galvanostat produced by Princeton Applied Research, USA (model PARSTAT 4000), and potentiodynamic polarization curves were plotted with VersaStudio software. Electrochemical tests were performed according to ASTM G5–94 (2011).

An electrochemical cell (Fig. 3) was used to perform the tests, which consists of a saturated calomel electrode (SCE) - reference electrode, a platinum electrode - recording electrode and the working electrode which consisted of the investigated samples.



Fig. 3 – The electrochemical cell

The evaluation of the corrosion resistance was performed by the polarization resistance technique. For this purpose, the open circuit potential (E_{oc}) was measured for a period of 6 hours and then the Tafel curves (potentiodynamic polarization curves) were drawn from -0.2 V (vs OCP)

to +0.2 V (vs OCP), using a scan rate of 0.167 mV / s.

Corrosion tests were performed at a temperature of $37 \pm 0.5^\circ \text{C}$ in a bath with heating and recirculation model CW-05G (Jeio Tech), in artificial saliva (SA) Carter Brugirard at pH = 7.58. (Table 3)

Table 3. Chemical composition of Carter Brugirard artificial saliva [25, 26]

No.	Substance	Quantity/L (g)
1	Na_2HPO_4	0.19
2	NaCl	0.7
3	KSCN	0.33
4	KH_2PO_4	0.26
5	NaHCO_3	1.5
6	Urea	1.3

Results

Figure 4 shows graphically the comparative variation of the open circuit potential (E_{oc}) for the samples investigated, and Figure 5 comparatively illustrates the Tafel curves for samples studied.

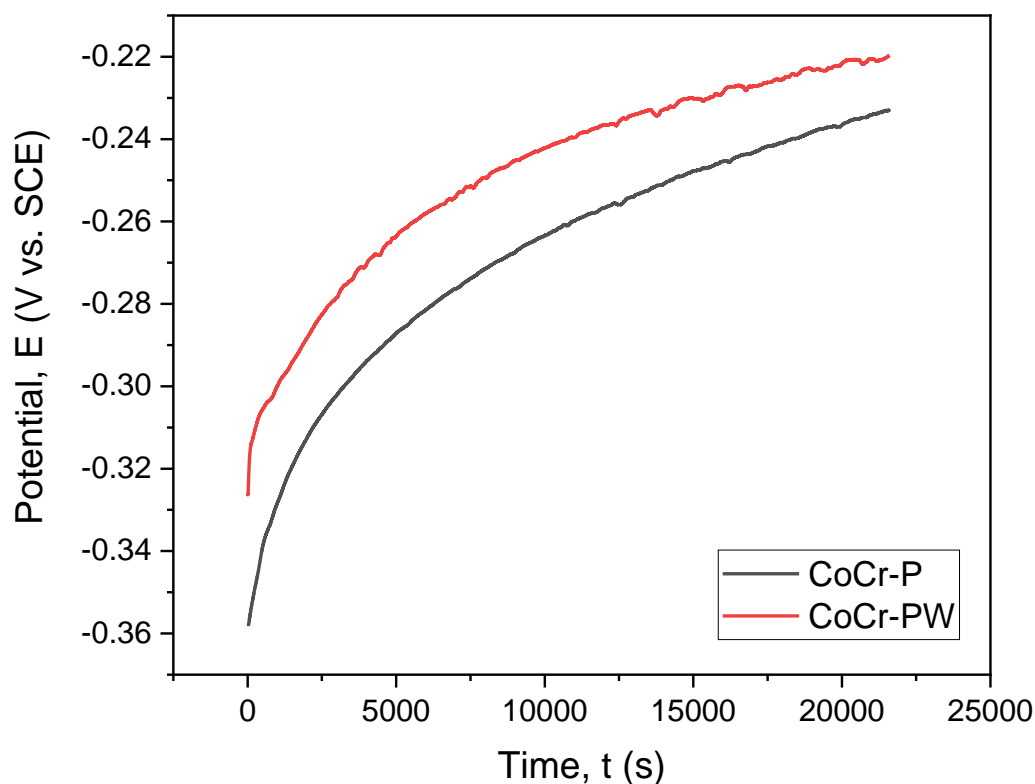


Fig. 4. Evolution of the open circuit potential (E_{oc}) for both samples tested

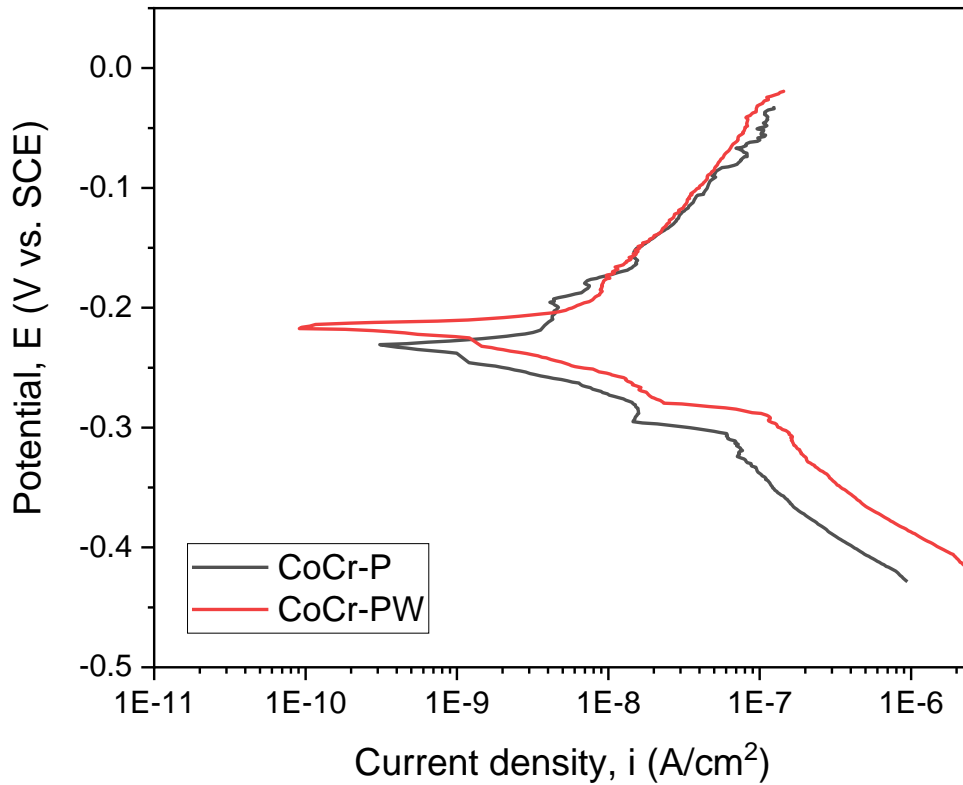


Fig. 5. Tafel curves for both samples tested

The parameters that characterize the corrosion resistance of the investigated alloys, namely E_{corr} (corrosion potential), E_{oc} (open circuit potential), i_{corr} (corrosion current density), β_a (anodic curve slope) and β_c (cathode curve slope), were determined by extrapolating Tafel.

Subsequently, we determined the polarization resistance (R_p) according to ASTM G59-97 (2014) using the equation:

$$R_p = \frac{1}{2.3} \frac{|\beta_a| |\beta_c|}{\beta_a + |\beta_c|} \frac{1}{i_{corr}}$$

where: β_a – anodic Tafel slope
 β_c – cathodic Tafel slope
 i_{corr} – corrosion current density

The corrosion rate was calculated according to ASTM G102-89 (2015) using the formula:

$$CR = K_i \frac{i_{corr}}{\rho} EW$$

where: CR – corrosion rate,
 $K_i - 3,27 \times 10^{-3}$,
 i_{corr} – corrosion current density
 ρ – material density,
 EW – equivalent weight.

The average values of the electrochemical parameters when the tests were carried out in artificial saliva for both studied Co-Cr based alloys are presented in Table 4.

Table.4 Main electrochemical parameters

Material	E_{oc} (mV)	E_{cor} (mV)	i_{cor} (nA/cm ²)	β_c (mV)	β_a (mV)	Rp (k Ω cm ²)	CR (nm/year)
CoCr-P	-233	-234	5,398	89,39	142,60	4425	57
CoCr-PW	-220	-221	5,503	62,91	153,86	3345	67

After performing the corrosion resistance tests, the tested surfaces were investigated from the point of view of the chemical composition with the aim of evaluating the oxide developed on the surface in terms of the amount of oxygen. The results obtained (Fig. 6) showed that oxygen was detected in a larger amount on each of the

tested alloys. In the case of CoCr-P alloy the amount of O increased from 2.42% to 3.75% representing an increase of about 55% and in the case of CoCr-PW alloy the increase was about 15% starting initially from 3.1 % and reaching, after corrosion tests, 3.57%.

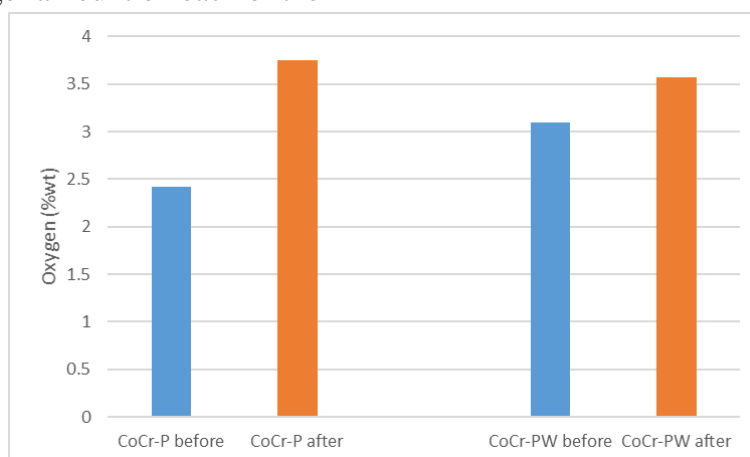


Fig. 6. Quantity of O quantified by EDS analysis before and after corrosion tests on investigated alloys

Discussions

As in other studies performed by the polarization resistance technique [27-31], the comparative evaluation of the corrosion resistance can be analyzed according to 5 criteria: open circuit potential, corrosion potential, corrosion current density, polarization resistance and corrosion rate.

The evaluation of the corrosion resistance of the two samples tested according to the value of the open circuit potential (E_{oc}) showed that the Co-Cr-PW test showed a more electropositive value and consequently a higher behavior, but it should be noted that the difference between the two E_{oc} values of the tested samples is not very high (13mV).

The comparative evaluation of the corrosion resistance in relation to the value of the corrosion potential (E_{corr}) showed that the Co-Cr-PW sample has the highest electropositive value of the corrosion potential (221 mV), as in the case of E_{oc} a difference of only 13 mV of the value recorded in the case of the probable Co-Cr-P.

The comparative evaluation of the two samples taking as a criterion the density of the corrosion current (i_{corr}), showed that the lowest value of the density of the corrosion current is registered in the Co-Cr-P sample (5.398 nA / cm²). However, the values of the corrosion current densities of the tested samples are quite close.

Taking the polarization resistance as a criterion for classifying the corrosion behavior, we find that the Co-Cr-P sample has the highest value (4425 k Ω xcm²), clearly superior to the Co-Cr-PW sample (3345 k Ω xcm²).

In terms of corrosion rate, the Co-Cr-P test showed the lowest value, 57 nm / year,

compared to the Co-Cr-PW test which recorded a corrosion rate of 67 nm / year in the tests performed.

Previous experimental studies on other Co-Cr or Ni-Cr alloys have helped to better understand their biocompatibility [32, 33], given that the oral environment is a corrosive environment that can adversely affect the integrity of prosthetic restorations through its saliva-containing structure. bacteria, possibly reflux gastric acid [34], but also by the temperature that fluctuates significantly when introducing cold or very hot foods and drinks [35-48].

Conclusion

Both tested alloys showed an increased corrosion resistance when immersed in artificial saliva, proving a good corrosion resistance and therefore a good biocompatibility. Comparatively analyzing the two tested alloys in terms of the 5 stated criteria (open circuit potential, corrosion potential, corrosion current density, polarization resistance and corrosion rate), the Co-Cr-P test showed superior electrochemical values and, implicitly, higher corrosion resistance.

The two alloys we studied have a similar chemical composition, but the corrosion rate is different because of the presence of Mo in the first one.

EDS analysis performed before and after corrosion test showed that the CoCr-P alloy has developed a thicker oxide layer on the surface than the CoCr-PW alloy, giving it better corrosion resistance, as confirmed by the results of corrosion tests in artificial saliva.

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