

RESEARCH AND EDUCATION

Cobalt-chromium alloys in dentistry: An evaluation of metal ion release



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Metal alloys used for fixed dental prostheses have been successfully used in clinical dentistry for long-lasting restorations.¹ Because they are in constant contact with mucosa, saliva, periodontal tissue, and bone, in-depth analyses of their chemical and physical characteristics and biocompatibility assays are essential to ensure patient safety. Biocompatibility is related to corrosion in a biological environment.² The release of metal ions during corrosion may lead to several possible consequences, including serious damage to patient health,³⁻⁷ allergies, oral lesions, or a salty or metallic taste.⁸ Metal ion concentration increases significantly in the saliva of patients with a metal dental prosthesis.⁹⁻¹² The oral environment is an ideal place for corrosion because of the presence of saliva, acid bacterial plaque, changes in pH and temperature related to food or beverage intake, and the action of different medications.¹³⁻¹⁷ The possibility of local alterations and the degree of severity at which they happen can be directly related to the alloy type and chemical state of

ABSTRACT

Statement of Problem. Metal ions released into the oral cavity from dental prosthesis alloys may damage the cellular metabolism or proliferation and cause hypersensitivity or allergies. The oral cavity environment is particularly prone to corrosion due to saliva, microorganisms, and pH variations.

Purpose. The purpose of this in vitro study was to evaluate the ion release of chromium, cobalt, and iron from the Co-Cr alloys used for traditionally cast and computer-aided design/computer-aided manufacturing dental devices after interaction with oral bacteria and different pH conditions.

Material and methods. All specimens were prepared from currently available alloys, polished, and immersed in 3 different pH media (artificial saliva [pH 2.3] and 6.5% and 0.9% saline solution [pH 7.1]). Specimens were also incubated in the presence of the bacterium *Eikenella corrodens*. Solutions were analyzed with an atomic absorption spectrometer after 15 and 30 days in the chemical corrosion test and 30 days in the biocorrosion test to detect ions released in different solutions. An ANOVA test was used to evaluate statistically significant differences among the percentages of metal corrosion ion release values.

Results. The greatest amount of element release was seen after 30 days: 4.964 ppm of casting alloy, 2.642 ppm of milling alloy, and 2.351 ppm of laser metal sintering.

Conclusions. With the exception of casting alloy under acidic conditions, no significant differences were found, even after exposure to bacteria. (J Prosthet Dent 2015;114:602-608)

the metal, the concentration, and the time of contact with mucosal tissues.^{4,5}

Base metal dental prostheses with monophasic alloys are less cytotoxic and inhibit human cell growth less than those with polyphasic alloys.^{9,10,18-20} Co-Cr alloys¹ seem to be the most reliable because the presence of Cr in amounts higher than 25 wt% provides good mechanical and biological properties and high corrosion resistance.²¹ Chromium on the order of 50 to 200 mg/day is an

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Clinical Implications

Co-Cr alloys are the base metal alloys used most for dental prostheses, but corrosion causes them to release ions dangerous to oral health. Because of its homogeneous microstructure and lack of porosity, a laser sintering alloy could replace traditional casting or milling alloys and better meet the needs of clinical applications with a low ion release.

important element for the human metabolism, whereas a lethal dose is approximately 50 to 70 mg/kg of body mass.²²⁻²⁶ Allergies to chromium have been reported in the general population, with average values between 3% (women) and 10% (men), although oral mucosa allergies are quite unusual.²⁷ Cobalt belongs to the prosthetic group of vitamin B12, which is essential for red blood cell formation, neurological function, and DNA synthesis.²⁸

The presence of specific bacteria, pathogens, or healthy commensal organisms in the oral cavity can trigger biocorrosion of dental alloys by activating processes not yet completely understood.^{3,28-30} Bacterial metabolic products, including organic and inorganic acid, affect the pH of the alloy surface, causing a change in composition of the medium that could promote development of corrosion. However, bacterial development of microbial corrosion on dental alloy requires the adhesion of bacteria to the surface, which also depends on characteristics.^{17,31}

Retention of oral microorganisms on cobalt-chromium alloys has been described, but the possible effect of specific bacteria on biocorrosion processes is still unclear.³² *Eikenella corrodens*, a facultative gram-negative bacillus, is a common inhabitant of the oral cavity.^{33,34} Its primary ecologic niche within the oral cavity appears to be the dental plaque in both healthy individuals and patients with periodontitis. It seems to play a role in biocorrosion processes as reported for others dental alloys.³⁵⁻³⁷ Co-Cr dental alloys can be prepared with different synthetic processes. The computer-aided design/computer-aided manufacturing technique is based on the milling of frameworks from a preformed block of alloy or by using direct laser metal sintering (LS), where a high-power laser fuses small metal alloy layers to reproduce the desired 3-dimensional object.^{3,15,38} These approaches are different from traditional casting techniques and appear promising for dental prostheses because they are more standardized and free of porosity defects that are detectable after wax casting.⁴⁰ The different microstructure and hardness described in other studies after preparation by different techniques suggest a different behavior when alloys are subjected to changes in pH or other oral microenvironment variations.^{19,40,41}

Milling alloys show an inhomogeneous microstructure consisting of a solid solution matrix interspersed with a second phase occupying the interdendritic spaces. In particular, the casting alloy (LW) revealed a high percentage of porosity because of interdendritic cavities created by shrinkage during the solidification of the alloy. This porosity is responsible for altering the strength and hardness of the prosthesis.⁴⁰ LS alloy, instead, has a homogeneous and compact structure, providing higher corrosion resistance and hardness.^{39,42}

The purpose of the present study was to evaluate the release of heavy metal ions from 3 differently fabricated cobalt-chromium dental alloys in the presence of saliva under physiological and altered conditions. The possible role of biocorrosion caused by oral bacteria was also investigated.

MATERIAL AND METHODS

Three different types of dental metal ceramic alloy were selected (all nominal compositions were provided by the manufacturer Ivoclar Vivadent): a casting alloy (60% Co, 30.1% Cr, 3.9% Ga, 3.2% Nb, < 1.0% Mo, Al, Si, Fe, B, Li); a milling alloy (61% Co, 28% Cr, 8.5% W, 1.65% Si, < 0.5% Fe, C); and a laser-sintering alloy (63.5 Co, 24.6% Cr, 5.1% Mo, 1.2% Si, < 0.50% Fe, Mn). All alloys were free of nickel, beryllium, and cadmium according to DIN EN ISO standard 22674:2011.⁴¹

Each dental alloy was shaped into 10 disks, each 15 mm in diameter and 1 mm thick. All specimens were polished with silicon carbide paper (400, 800, 1200 grit) with a grinder-polisher machine (M2V; Manfredi). The specimens were then cleaned in ethanol and deionized water in an ultrasonic cleaner for 5 minutes to remove contaminants and finally autoclave-sterilized as previously reported by Xin et al⁴² and McGinley et al.¹⁷

Three solutions were prepared in order to evaluate chemical corrosion following the study by Mutlu-Sagesen et al³⁰ and according to ISO standard 10271⁴³: 0.9% NaCl saline solution (SS) at pH 7.1; an artificial saliva solution (AS-LA) with 10^{-2} M lactic acid (pH 2.3) added; and an artificial saliva solution (AS-SS) with 0.252 g/L NaOH (pH 6.5) added. Artificial saliva is composed of $2.0 \cdot 10^{-2}$ M KCl, $1.4 \cdot 10^{-3}$ M KH_2PO_4 , and $1.5 \cdot 10^{-2}$ M K_2HCO_3 .

Three solutions were prepared to evaluate biocorrosion: a solution of 50% artificial saliva (pH 7.1), 45% tryptic soy broth (TSB; Oxoid) and 5% defibrinated horse blood (SS+B; Oxoid); a solution of 50% artificial saliva buffered with H_3PO_4 at pH 5.5, 45% TSB (Sigma-Aldrich) and 5% defibrinated horse blood (SS+AB; Oxoid); and a solution of 50% artificial saliva (pH 7.1), 45% TSB (Oxoid), and 5% defibrinated horse blood (Oxoid) added with 5×10^6 cells/mL *E. corrodens* (SS+BB). One sample of artificial saliva/TSB blood was adopted as blank reference. The 3 alloys were incubated in the

presence or absence of bacteria. The experiment was conducted in parallel with all solutions, SS+B, SS+AB, and SS+BB. Because of the slow growth of *E. corrodens*, the incubation time was extended up to 1 month, and the bacterial load was confirmed by using microscopy.

Each metal ceramic alloy was immersed in the solutions in polypropylene test tubes and incubated at 37°C to simulate the oral temperature. The solutions were analyzed after 15 and 30 days in the chemical corrosion test and 30 days in the biocorrosion test to detect ions released in different solutions. The biocorrosion test was conducted in a controlled atmosphere with 5% CO₂ concentration.

Solutions were then analyzed with an atomic absorption spectrometer (AA600; Perkin-Elmer) in a graphite oven (HGA500; Perkin-Elmer). During the analysis, the argon flow was 200 mL/min; the sample volume, increased, temperatures, and drying and atomization times were optimized before the analysis to achieve the maximum absorbance and the minimum background. Mg(NO₃)₂ (Merck) was used as a modifier according to the Perkin-Elmer manual.

Relative standard deviation (SD) and SD percentage (%RSD) calculations were based on 5 replicate specimens. The same analyses were conducted with certified control specimens (CRM-TMDW) with a Co recovery value of 96.1%, Cr of 97.5%, and Fe of 95.9%. The limit of quantification (LOQ) is defined as a level above which quantitative results can be obtained with a specified degree of confidence. At the 95% confidence level, the recommended LOQ value is 10 SD/m and the obtained value of LOQ for the adopted analytical method was 0.3 µg/L. The detection limit of each element was calculated as 3 times the SD of the blank (3σ blank, n=5 specimens); following the GF-AAS method, the detection limit for Cr was calculated as 0.003 mg/L, 0.0061 mg/L for Co, and 0.004 mg/L for Fe.

A 2-way ANOVA was used to evaluate statistically significant differences among the percentage of metal corrosion ions released and to evaluate the effect of the contact period with the different solutions. Differences between the averages of the factors were evaluated by the Fisher least significant difference test ($\alpha=.05$).

Metal ion concentrations were normalized relative to that of the blank (without dental alloy). For each alloy, the release of all metal ions in the different solutions (artificial saliva, artificial saliva with phosphoric acid, artificial saliva with *E. corrodens*) was evaluated and compared with 1-way ANOVA (MiniTab 16; Minitab Inc) to determine whether they differed significantly.

RESULTS

The results of metal ion (Cr, Co, Fe) release from the different alloys incubated in AS-SS, SS, and AS-LA

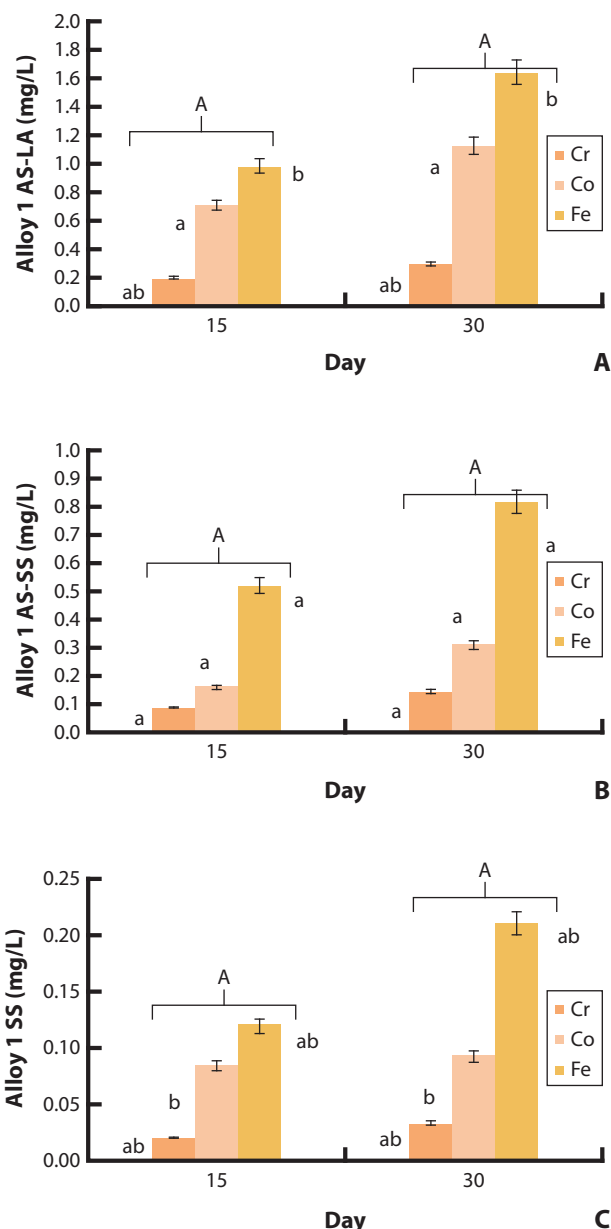


Figure 1. Release of Co, Cr, and Fe (mg/L) from LW. A, AS-LA solution. B, AS-SS solution. C, SS solution. Error bars = 1 SD of n=5 specimens. Uppercase letters indicate statistically significant differences in amounts released from the immersion group between the periods ($P<.05$). Lowercase letters indicate statistically significant differences among immersion solutions in each time period ($P<.05$). AS-LA, artificial saliva solution; AS-SS, artificial saliva solution added; LW, casting alloy; SS, saline solution.

solutions are shown in Figures 1-3. Differences were found in the release of chromium, cobalt, and iron ions between the successive time intervals (15 and 30 days) in all solutions; however, the ion release from all alloys was low, sometimes at the limit of detection. The results for pH 2.3 were obtained by using a 10⁻² M lactic acid solution and pH 6.5 with 0.252 g/L NaOH (Fig. 1). The pH

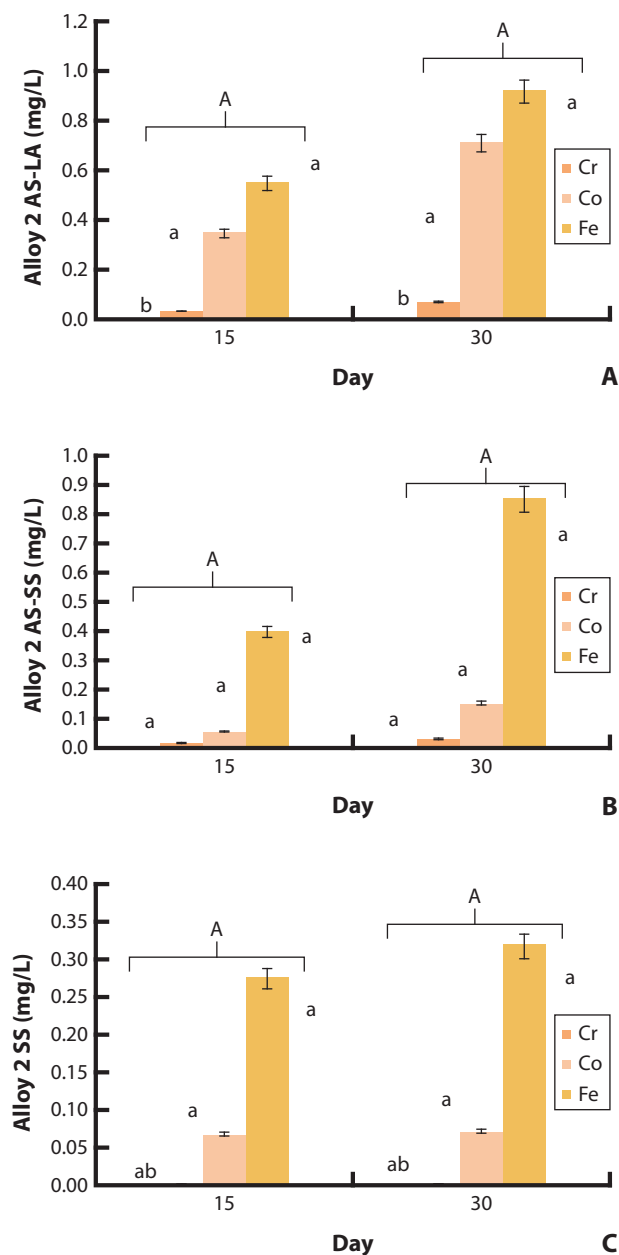


Figure 2. Release of Co, Cr, and Fe elements from M alloy. A, AS-LA solution. B, AS-SS solution. C, SS solution. Error bars = 1 SD of $n=5$ specimens. Uppercase letters indicate statistically significant differences in amounts released from the immersion group between the considered periods ($P<.05$). Lowercase letters indicate statistically significant differences among immersion solutions in each time period ($P<.05$).

effect on the quantity of elements released from the alloy produced by the casting synthesis method (LW) decreased linearly with increasing pH. The pH effect on the quantity of Cr released from the LS alloy was small. However, the quantity of Cr released from these alloys was relatively small in all the solutions (Fig. 3). The mean

value for the release of metals of the 3 alloys in different solutions during the 1 month of the experiment together with the 2-way ANOVA results are shown in Figures 1-3.

The correlation between metal concentrations and immersion time was generally poor. For immersion time, the release of all metals did not present statistically significant differences among the alloys ($P>.05$). The pH of the solution showed a statistically significant effect on the release amount ($P<.05$). The kind of alloy, the solution type, and the interaction of these 2 factors had a statistically significant influence on the Cr, Fe, and Co released from the alloy produced by the casting synthesis method (LW) and on the Cr released from alloy produced by the milling synthesis method (M) ($P<.05$). In the alloy produced by the milling synthesis method (M), significant differences were found in the determination of Cr in the acid and saline solution, but the release of Cr was at the limit of detection. For all dental alloys, the release of metal ions showed no significant differences under the different conditions tested.

DISCUSSION

Knowledge of the degree of metal ion release is important in order to prevent possible adverse effects for the patient such as toxicity, metallic taste, mucositis, gingival hyperplasia, or gingivitis.^{34,36} Moreover, the oral microflora plays a relevant role because the presence of specific bacteria, pathogens, or healthy commensal organisms can trigger biocorrosion mechanisms on dental alloys, following processes that are not yet completely understood.^{7,30}

Several factors can change the properties of a dental metal ceramic alloy, including casting and heating and cooling processes during preparation, or the porcelain firing process.¹² These phases may alter the metallurgical state, surface oxides, and corrosion properties of the alloy depending on its chemical composition. Autoclaving, in contrast, has no effect on the surface characteristics or chemical property of an alloy because of the low temperature reached (120°C to 160°C) in steam sterilization compared with the melting temperature of base dental alloys (1000°C to 1400°C).¹⁷

A multiphase (heterogeneous) alloy is more prone to corrosion than a monophasic or single-phase (homogeneous) alloy because of a galvanic effect between areas of different compositions inside the alloy.^{18,19,29} Can et al²⁰ emphasized that the release of metal ions from dental cast alloys has correlated with the surrounding environmental conditions and the surface of dental alloy. Because the use of different alloys and manufacturing processes may influence the properties of the final restorations, it is important to consider standard materials and established protocols when comparing research data.²⁹ Because metal ion release can be influenced by the composition, microstructure, and surface properties

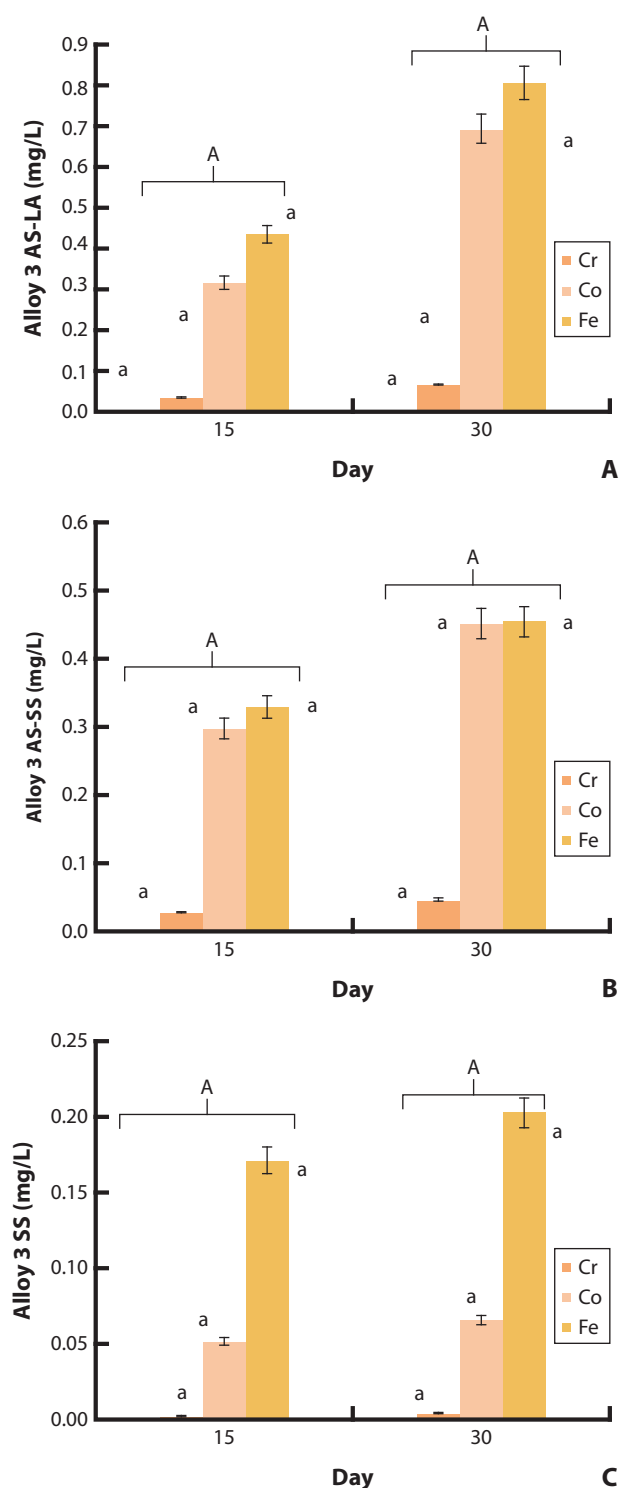


Figure 3. Release of Co, Cr, and Fe elements from LS. A, AS-LA solution. B, AS-SS solution. C, SS solution. Error bar indicates 1 standard deviation of $n=5$ specimens. Uppercase letters indicate statistically significant differences in amounts released from the immersion group between the periods ($P<.05$). Lowercase letters indicate statistically significant differences among immersion solutions in each time period ($P<.05$).

of the alloy, depending on the manufacturing technique used,⁴⁰ special attention should be paid to selecting the manufacturing method.

Using x-ray scanning, Al Jabari et al³⁹ found gross porosity only in dental alloys fabricated by means of conventional casting (LW). The interdendritic porosity due to shrinkage during the alloy solidification weakens the structure of the prosthesis. Grains of microstructure are not well defined and consist of few large crystals with each crystal forming a simple dendrite.⁴¹ No porosity was detected in milling (M) or laser sintering (LS) alloys. Moreover, different microstructures were identified among the groups by scanning electron microscopes (SEM) examination, energy dispersive using x-ray and x-ray diffraction analysis. Both cast and milled specimens showed an inhomogeneous structure with a dendritic matrix and a dispersed second phase that occupies the interdendritic spaces in their matrix. Precipitates of a third phase were also detected in the milled specimens. Such a phase was absent in the LS specimens, which did not show dendritic-like structural design. The absence of a second Mo-rich phase seems to eliminate the possibility of galvanic coupling within the alloy phase.⁴⁰

The LW group showed higher hardness than the milled group because of finer distribution of the disperse phase. The LS showed the highest value because of the absence of porosity and the more fine-grained structure attributed to the sintering technique.^{25,26,39,40}

Collected data suggested that for the LW alloy, the ion release was significantly higher ($P<.05$) in artificial saliva of pH 2.3 and lowest in saline solution, in agreement with findings by Mutlu-Sagesen et al.²⁹ However, the ions released from all the alloy was very low for all the studied solutions and exposure conditions. These observations could be explained by the anticorrosion properties of Co-Cr alloy, as related to the formation of the spontaneous oxide layer on the surface of the alloy when it is exposed to atmospheric oxygen after polishing.²⁶ Xin et al²⁶ examined the electrochemical corrosion in artificial saliva of LS and LW and found the oxide layer thickness on the cast specimens was 3.3 nm and 3.6 nm on the LS specimens, and the amounts of Co and Cr on respective specimen surfaces were similar.⁴²

Moreover, this study demonstrated that the iron ion was released at a higher level in the first period and then progressively diminished in all solutions, consistent with reports by Huang et al¹² and Kuhta et al,¹³ but in contrast to that by Okazaki et al.¹⁴ The higher level of the release of iron ion compared with chromium and cobalt seems not to comply with the presence of these elements in the alloy composition.

No significant differences in ion release could be revealed when the dental devices were exposed to a bacteria-enriched environment. This result only partially agrees with those of similar experiments performed with

the bacterium *Streptococcus mutans*, a common inhabitant of the oral cavity.¹⁵ One reason for this discrepancy can be found in the different biological properties of *E. corrodens*. This microorganism is described as the etiological agent of several oral inflammation processes and is a facultative anaerobic bacillus able to form biofilm; however, its ability to create an acidic environment in the mouth has not been verified.^{16,17} Probably *E. corrodens* is not able to trigger acid biocorrosion on the investigated alloy. This hypothesis is supported by our data showing that different metal ions are significantly released after exposure to a chemical solution with low pH (pH 2.3) values for all the tested alloys.

The hypothetical biocorrosion effect could have been reduced also by the absence of a complex biofilm on the alloy surface, comparable with oral plaque and its microflora. *E. corrodens* has indeed been found in human dental plaque in association with other prokaryotic species.³² *E. corrodens* can play a role in specific co-aggregation with certain gram-positive and gram-negative bacteria present in human periodontal pockets but probably cannot produce a stable biofilm without interaction with other microflora components.^{31,34,35,37}

The final aim of this study was not to reproduce a biofilm comparable with dental plaque but to detect a possible effect of a highly abundant oral prokaryotic species. However, the biological effect investigated in this study could have been reduced by the simplified in vitro model. Even if managing microflora dynamics is challenging, future experimental models should consider the use of a bacterial mixture to mimic human oral plaque and its complex interactions with alloys.

CONCLUSIONS

Co-Cr alloys processed with different fabrication techniques (casting, milling, and laser sintering) show a release of low amounts of ions in all tested conditions. No difference was noted when the specimens were exposed to an enriched bacteria environment. It was concluded that all the alloys are adequately corrosion resistant and well suited for dental usage, although the LW alloy shows the greatest risk of corrosion under acidic conditions.

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