Some consequences of repeated casting of Co–Cr dental alloy

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Abstract: The effect of multiple recasting of commercial Co–Cr base dental alloy (Dentalite-C) on its structural characteristics and ion release rates in artificial saliva has been investigated. The number of recasting was once, four and eight times. The as-cast microstructure became uniformer as the number of recasting was increased whereby macro- and micro-hardness and electrical conductivity of the castings decreased. Metallic ion release in artificial saliva was monitored *in vitro* over the period of 22 weeks at the temperature of 37 °C and pH 2.3. Only Co and Fe were evidenced in artificial saliva and their release rates are measured. The ion release rates increases with time. The ion release rate of Fe was larger than Co. The ion release rates increases substantially by increasing the number of recasting.

Keywords: Co-base alloy; recasting; macro- and micro-hardness; artificial saliva; metallic ion release rate.

INTRODUCTION

Non-precious (base) dental alloys are rapidly coming into the center of attraction replacing precious or noble alloys in dentistry. They are being used in full-cast and metal–ceramic restorations besides removable partial dentures. Approximately 90% of all removable partial dentures are now cast from non-precious alloys containing Co, Cr and Ni.[1] Co–Cr alloys were first applied in dentistry in 1929. Besides their economic advantage over gold, these alloys are also attractive because they have less than half the density and are considerably stronger than gold.[2] These alloys are not without their disadvantages. They are more difficult to cast than non-base metal alloys (high noble and noble alloys) because of their very high liquidus temperatures. They also exhibit a greater casting shrinkage (about 2.3%) that must be compensated for. Due to their lower

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ductility and greater hardness, they are less burnishable and more difficult to finish and polish than the high noble and noble metal alloys.

Currently, the majority of commercial dental laboratories are reusing scrap alloy (used once or more times) either alone or in combination with new alloy for fabrication the dental prosthesis. Actually, they consider only the cost benefit obtained using scrap alloy, without any regard to manufacturer's instructions or alterations of properties as a consequence of reusage.[3] It is well known that recasting of base metal alloys changes the chemical properties of the material and may be the cause of the element release.[4,5] According to clinical implications, recasting of base metal alloys markedly increases their potential cytotoxic effect. Thus, using this procedure in the dental practice may adversely affect the soft tissue adjacent to a restoration. Therefore, recasting of the base metal alloys should be avoided in the clinical dentistry.[6] Identification and quantification of the released elements is the most relevant measure of bio-compatibility.[7] Ion release from dental alloys has been evaluated mainly by *in vitro* studies using the following methods: cell culture media,[8–14] galvanism,[15] electrolyte baths,[16–18] evaluation of the influence of oral proteins on corrosion behavior[19] use of different surface treatments to cast alloys,[20,21] exposure to different pH levels[7,10,22,23] and power tooth brushing with toothpastes of different abrasive-ness.[24] In living systems, some metals have biological functions. While Fe is essential in relatively high concentrations, other elements such as Co, Mo, Cu and perhaps Cr, are essential only in trace amounts. However, at higher concentrations, they can become very toxic.

When an alloy is placed in the oral environment, interaction between alloy and saliva takes place. The effect of this interaction may be manifested as: *i*) release of soluble metallic ions, *ii*) retaining of corrosion products on the alloy surface or *iii*) a combination of both. Depending on the altered alloy surface and/or the nature of released metallic ions, these effects may trigger adverse biological reactions such as allergy.[25] The pH of saliva may vary between 2 and 11 while the temperature in the oral cavity may be between 0 and 70 °C, all these variations mostly depend on the food intake. Human saliva is a complex fluid secreted by the salivary glands and its normal pH is slightly alkaline (pH 7.4). Saliva contains organic and inorganic substances suspended in an aqueous medium.[1]

There are a large number of papers dealing with the release of ions from various types of dental alloys in artificial saliva under different conditions (pH, composition, temperature, *etc.*). However, the authors followed the ion release over a period between 7 and 30 days, as prescribed by some standards, but without details on the kinetics of the process.[7,10–12,22–27]

Due to possible material savings in making dental appliances, there is a possibility to use multiple casted dental alloys. Multi-casting may be regarded as a

kind of stress that violates the original structural properties of the alloy, provoking aging processes that increase the possibility of material failure and, especially, risk to human health. The goal of this study was to investigate the effects of multiple recasting on Co–Cr base dental alloy (Dentalite-C). Recasting was performed once, four times and eight times. Alterations in the alloy microstructure, macro- and micro-hardness and electrical conductivity were followed. Specifically, a long-term monitoring (22 weeks) of the ion release in artificial saliva at the temperature of the human body was emphasized in order to measure the release rates. The release rates of Fe and Co in all three casts were quantified and correlated with the number of recastings.

EXPERIMENTAL

Materials and methods

Dentalite-C, a commercially available Co–Cr base dental alloy (Sanitaria, Austria), was used in this study. The declared nominal composition as well as the compositions determined by inductively coupled plasma optical emission spectrometry (ICP-OES, Spectroflame, 27.12 MHz and 2.5 kW, model P) and X-ray fluorescence spectrometry (XRF, Thermo Scientific Niton XL3t 980 Gold+) are summarized in Table I**.**

TABLE I. Elemental composition of the Dentalite-C alloy used in this study

Element	Declared, wt. %	ICP-OES, wt. %	XRF, wt. %
Co	60	59±2	62.2±0.9
Cr	30	23.0±0.4	27±1
Mo	5.5	5.9±0.2	6.0±0.1
Nb	1	0.84±0.01	1.5±0.1
Fe	Remaining	0.92±0.03	1.06±0.03
Mn	Remaining	0.346±0.004	0.3±0.1
Cu	Remaining	0.35±0.01	0.53±0.02
Si	Remaining	0.095±0.003	0.79±0.05

Production of test samples – casting

Test samples were disk-like castings obtained by melting in an induction furnace. In accordance with the manufacturer's instruction, melting was performed at temperature 1450 °C. Multiple recast samples were obtained by successive melting and casting of the same sample. Dimensions of samples used in the ion release experiments are presented in Table II. Diameter and thickness were measured with 0.01 mm accuracy and weighed with 0.1 mg accuracy. The densities were consequently calculated and the declared density is 8.2 g cm^{-3}. Precision is given as the standard deviation of five measurements. The dimensions of the samples used for other measurements were similar to these.

TABLE II. The dimensions of the cast samples used for testing

Cast	Diameter, mm	Thickness, mm	Surface[a], mm^2	m / mg	V / mm^3	ρ / g cm^{-3}
1	9.85±0.05	1.06±0.01	185±2	620.6±0.1	81±3	7.7±0.3
4	9.94±0.14	1.55±0.04	204±2	895.0±0.1	120±8	7.5±0.5
8	9.89±0.07	0.83±0.05	179±2	456.4±0.1	64±5	7.1±0.6

[a]Both surfaces are given

Sample pre-treatment

After casting, the test samples were primarily wet ground with silicon carbide papers down to 2000 and subsequently polished with diamond paste with 3 µm and 1 µm particle size. Finally, the samples were polished with 0.05 µm alumina slurry and then washed ultrasonically in water and ethanol and air-dried. All tests were performed with pre-treated samples.

Characterization of microstructure

Test samples were additionally etched for a 20 s in a mixture of 15 mL distilled water, 15 mL glacial acetic acid, 60 mL hydrochloric acid (32 %) and 15 mL nitric acid (65 %).[28] Samples treated in this way were examined by scanning electron microscopy (SEM, JEOL--JSM 5800LV) and FEI Quanta 200 equipped with an energy dispersive spectrometer (EDS) (Oxford Instruments INCA X-sight system). The chemical composition of dendrite and inter-dendrite regions was determined by point and bulk surface analyses.

Hardness testing

Macro- and micro-hardness tests were performed using a Buehler SemiMacroVickers 5112 HV112 and a Buehler MicroMet 5101 Vickers testers. The applied loads and times of indentation were as follows: 1 kg for 5 s and 25 g for 10 s, respectively. The results are presented as an average of five measurements.

Electrical conductivity testing

The electrical conductivity was measured using a conductivity meter (Foersted Sigmatest 2.069).[29] A frequency of 960 kHz was applied due to the small thickness of samples. The results are presented as an average of ten consecutive measurements.

Elemental analysis

Elemental analysis of cast samples as well as concentration of metal released in artificial saliva was performed by ICP-OES. A small piece of a cast sample was weighed with an accuracy of 0.1 mg, poured with 10 mL of 1:1 HCl (Fisher Chemicals) and repeatedly heated until complete dissolution. Certified J.T. Baker standards were used to prepare standard solutions in µg mL^{-1} and sub µg mL^{-1} ranges of the constitutive elements. For the case of metal concentration determination in saliva solutions, the standards were matched to the saliva matrix. Limits of quantification for the base elements were between 0.05 and 0.15 µg mL^{-1}. Samples were measured at least in triplicate and the average value is presented. Deionized water was used for all preparations (Purite Ltd., Oxon, UK).

Procedure for the measurement of the metallic ion release rate

The choice to use artificial saliva as the electrolyte at open atmosphere and a temperature of 37 °C was made with the intention of having conditions as similar as possible to clinical conditions. An artificial saliva solution of pH 2.3±0.1 was prepared as follows: 7.69 g K$_2$HPO$_4$, 2.46 g KH$_2$PO$_4$, 5.3 g NaCl, 9.3 g KCl (Honeywell Riedel-de Haen), and 16.6 mL 90 % lactic acid (Fluka Chemica) were dissolved into 1 dm^3 of deionized water.[10]

One sample of each kind of casting was put into an Erlenmeyer flask containing 250 mL of artificial saliva solution and closed. The solutions were permanently stirred at moderate speed whereby the sample remained stationary on the bottom. The temperature was set at 37±1 °C and controlled on a daily basis. From each flask, aliquots of 25 mL were sampled every seven days during 10 weeks and stored for later elemental analysis. The last aliquot had

been left for 22 weeks (154 days), but without temperature control. During this period temperature varied on a daily basis between 18–25 °C.

This regulated sampling scheme required correction of metal concentrations determined in successive aliquots. Due to the progressive volume reduction, the actual metal content should be re-calculated relative to the starting volume (250 mL), *i.e.*, to be multiplied by the quotient $(250–x·25)/250$, where $x = 0,1,2,…,8$ represents the aliquot sequence. That is comparable to ten individual aliquots of 250 ml of artificial saliva each of which contained one piece of sample. Finally, metal concentrations should be normalized onto the mass ratio of the constitutive element in the specimen used.

Such experimental design was verified in that the four times recast sample was treated slightly differently. Namely, it was immersed into 200 mL of artificial saliva and nine aliquots, each of 20 mL were sampled during 15 weeks. The timing of sampling was not uniform, but varied between 5 to 20 days. The temperature was controlled throughout the 15 weeks.

RESULTS AND DISCUSSION

Microstructure and hardness

The microstructures from the center of the as-cast samples after the first (a), fourth (b) and eighth recasting (c) are shown in Fig. 1a–c, respectively. All samples exhibited a solid solution matrix with a typical dendritic arrangement. It is well known that a dendrite structure exhibits compositional variations, with the dendritic arms containing less alloying and impurity elements than the interdendritic regions. Due to these compositional changes, the rate of etching of interdendritic regions differs from that at the dendritic arms. There were noticeable changes in the microstructure with the number of recasting. The SEM microstructure of the once-cast sample consisted of a dendritic matrix with precipitates present at the grain boundaries and interdendritic regions (Fig. 1a). This is in agreement with literature results reported for these types of alloys in the as-cast condition.[30,31] With the increasing the number of recasting, the clear distinction between the matrix and dendrites disappeared and the microstructure became more homogeneous (Fig. 1b and c).
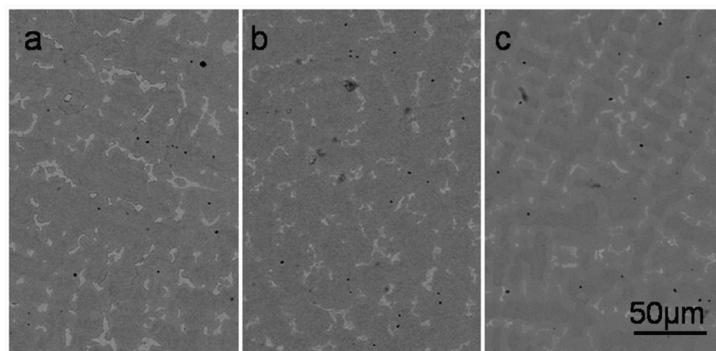

Fig. 1. SEM (BSE) images of Co–Cr dental alloy. Effect of number of recastings on the microstructure of the alloy: (a) once casted, (b) four times and (c) eight times casted.

EDS analysis confirms that the distribution of the base elements into dendritic and interdendritic regions were different depending on the number of recastings. In the once-cast sample, the amount of Co in the interdendritic regions was about 20 % less in comparison to the bulk sample. In contrast, the amount of Mo was much higher (about 2.5 times) in the interdentritic regions, (Fig. 2). The amount of Cr was always homogenously distributed in both regions and was independent on the number of recasting (the average concentration ratio for all three kinds of samples was 0.97±0.08).
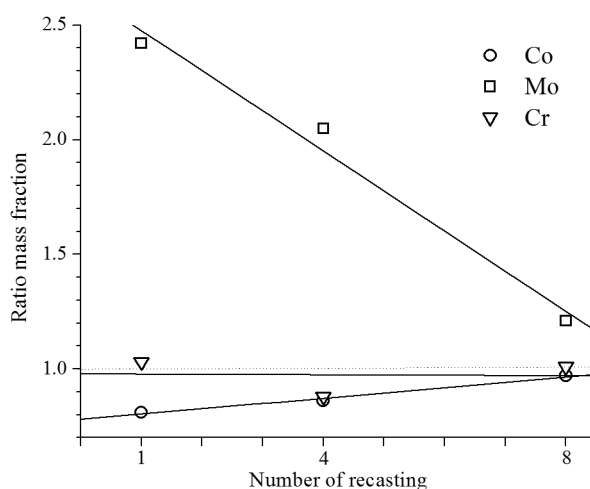

Fig. 2. Concentration ratios, ratio of the mass fraction of Co, Mo and Cr in the interdendritic region to the mass ratios in a bulk piece as a function of the number of recastings.

With increasing number of recastings, the distribution of the Mo and the Co contents into the dendritic and interdendritic regions tended to equalize, (Fig. 2). It was expected that after a certain number of recasting their contents would become uniformly distributed.

Correlation between the number of recastings and the macro- and micro-hardness of the dendritic and interdendritic region of the alloy is shown in Fig. 3. It may be seen that with increasing number of recastings, the macro- and micro-hardness decreased and the micro-hardness of the interdendritic regions was higher than that of the dendritic areas. The reason for such a difference may be ascribed to the change in the ratio of the alloying elements, which is in accordance with the results of EDS analysis, Fig. 2. The difference between the mircro-hardness of the dendritic and interdendritic regions decreased following the sequence (150→140→110) MPa. The macro- and micro-hardness measurements of individual regions (dendrite, interdendritic region) in the case of the sample after the first casting can be seen in Fig. 4. Several indents were made in the

primary dendrite region, which had an average hardness of 253 MPa. The inter-dendritic region had an average hardness of 402 MPa. The same measurements were performed on the other samples. The average macro-hardness of the sample subjected to only one casting reached a value similar to that declared by the manufacturer, *i.e.*, 328 MPa.
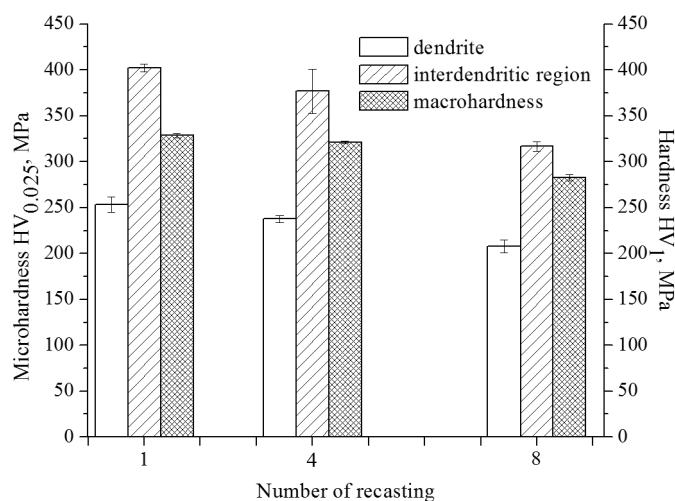

Fig. 3. The effect of the number of recastings of Co–Cr dental alloy on the micro-hardness of dendritic and interdendritic regions and the macro-hardness of bulk samples.
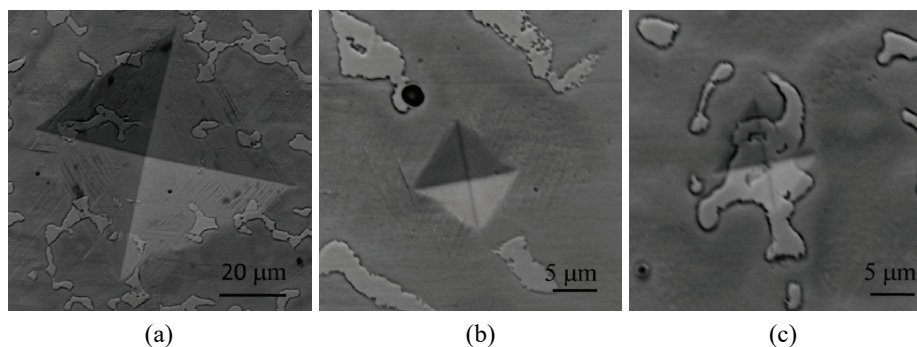

Figure 4. Macro-hardness (a) and micro-hardness measurements of individual region dendrites (b) and interdendritic region (c) in once casted sample; SEM (SE) images of selected regions.

Recasting comprises remelting and resolidification and the new grain size may be different from the original one. When the melting temperature and casting conditions are the same (as in this case), the new grain size depends on the concentration of nuclei for grain formation.[32] Generally, with each recasting, the impurities may facilitate grain nucleation and decrease the grain size inducing an increase in the macro-hardness. However, in the present study, the macro- and

micro-hardness slightly decreased with increasing number of recastings. It could be concluded that this property depended much more on the structure type of the phases than on the difference in their chemical composition and the grain size.[33]

Electrical conductivity

Information about electrical conductivity for most of the materials used in dentistry is not readily available. Starčuková *et al.*[34] reported that the differences in the electrical conductivity of 45 metallic dental materials are in the range from 0.8 to 9.41 MS m^{-1}. The electrical conductivity of cobalt–chromium alloys was found to be low and in the narrow range of 1.08–1.26 MS m^{-1}. The values of electrical conductivity of recast samples of Co–Cr based dental alloy indicated that with increasing number of recastings, the concentration of impurities, such as oxides, nitrides and carbides, was increased, which caused the electrical conductivity to decrease, Fig. 5.
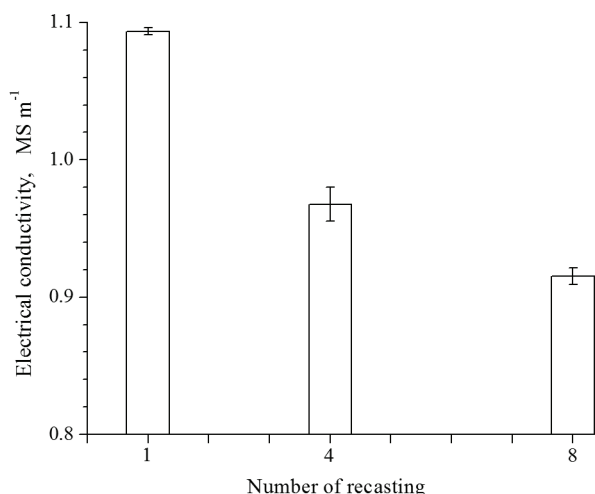

Fig. 5. Electrical conductivity of multiple casted samples of Co–Cr dental alloy.

Ion release rate

Metallic ion release was monitored over a period of 22 weeks. Of all the constitutive elements of the alloy, only Co and Fe were detected in the artificial saliva and could be considered. Other basic as well as concomitant elements were below the limit of quantification.

Cumulative increase of Fe and Co concentration in saliva fits well a two-parameter Weibull cumulative distribution function (CDF):

$$F(x) = 1 - e^{-(x/n)^k} \tag{1}$$

where $n > 0$ and $k > 0$ correspond to the scale and shape parameter, respectively, and x > 0.

Due to its flexible shape and ability to model a wide range of random phenomena, the Weibull CDF was used successfully in many applications as a purely empirical model. It has the potential to describe microbial, enzymatic and chemical degradation kinetics subjected to stress conditions.[35] In addition, Weibull statistics is often used in the analysis of data addressing mechanical properties (fatigue) of various materials, including dental metal and ceramic materials.[36,37] Hence, the scale parameter, n, is often referred to as a characteristic life, or failure rate.[37] The scale parameter identifies the elapsed time of the entire process being monitored, and hence when $x = n$, then $F(x) = 1-e^{-1} \approx 0.63$, meaning 63 % of the entire process has been finished. Depending on the value of the shape parameter k, the Weibull distribution can describe three types of time-dependent processes. If the shape parameter $k = 1$, the Weibull distribution reduces into an exponential that describes the process occurring at a constant rate. If the shape parameter $k > 1$, the Weibull distribution describes a process that accelerates in time. Finally, if $0 < k < 1$, then the process decelerates.

In practice, the Weibull distribution parameters are calculated by the linear regression method, Eq. (2). After rearranging Eq. (1), the linear form is obtained:

$$\ln\{-\ln(1-F(x))\} = k \ln x - k \ln n \qquad (2)$$

where k corresponds to the slope, and $k \ln n$ to the intercept of the linear fit. Thereby, $F(x)$ corresponds to the fractional cumulative concentration of a metallic ion in solution over time x (weeks in this study). The linear fit plots for Fe and Co in the case of the eight times recast sample are shown in Fig. 6. The last
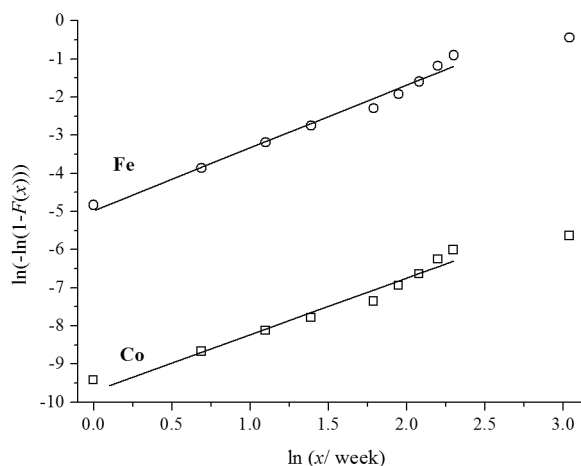

Fig. 6. The Weibull linear fit plots for Fe and Co in the case of the eight-time casted sample.

points (22 weeks) deviates downwards and was not included in the linear regression. The deviations originate from the temperature reduction of the saliva in the period of the 10th to the 22nd week.

The linear fit plots for Fe in the cases of once, four times and eight times recast samples are shown in Fig. 7. In the cases of once and eight times casted samples, the last points (22nd week) deviates downwards and is not included in the linear regression. In the case of four times cast sample, the regression is linear up to 15 weeks since the temperature of the saliva was kept at 37 °C.
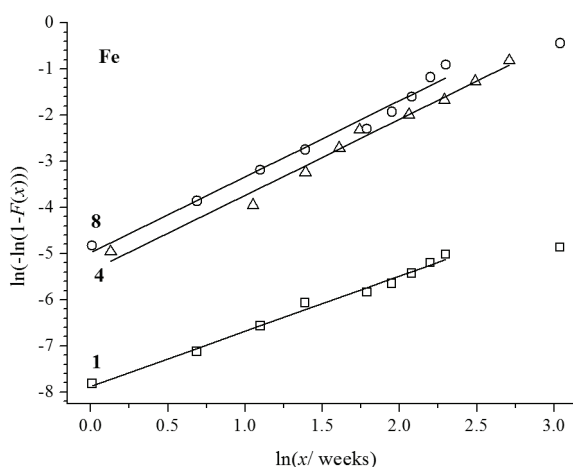

Fig. 7. The Weibull linear fit plots for Fe for once, four times and eight times casted samples.

Parameters values of the Weibull distribution for both elements and three classes of the casts are summarized in Table III. In all classes of casts, the shape parameter was k > 1, which means the release rate of Fe and Co ions in saliva increased over time. The only exception could be in the case of Co and for once cast sample for which the shape parameter $k \approx 1$, Table III. Taking into account the error of the regression analysis, the release rate actually increased slowly,

TABLE III. Parameters of the Weibull distribution linear regression, Eq. (2), and corresponding scale parameters for Fe and Co in all three classes of casts

Ion	No. of recasts	k	$-k\ln n$	n, weeks	R^2
Fe	1	1.20±0.04	7.88±0.07	711±38 (≈13.7 year)	0.98912
	4	1.66±0.07	5.5±0.1	27±2 (≈0.5 year)	0.98475
	8	1.65±0.08	5.0±0.1	21±2 (≈0.4 year)	0.97994
	∞	–	–	11±1 (≈0.2 year)	–
Co	1	1.10±0.06	11.3±0.1	28930±3000 (≈556 year)	0.97252
	4	1.58±0.05	9.5±0.1	409±10 (≈8 year)	0.99122
	8	1.5±0.1	8.6±0.2	310±80 (≈6 year)	0.98429
	∞	–	–	134 (≈2.5 years)	–

which confirmed the remarkable long scale factor for Co. The release rates of Fe were notably higher than those of Co, although Co is the base element of the alloy, Table III.

The values of the scale parameter show that the release rates of Fe and Co rapidly increased with increasing number of recasts (Table III). This is in correlation with the changes in the mechanical characteristics (macro- and micro-hardness, Fig. 3, and electrical conductivity, Fig. 5. Successive melting and casting equalizes the chemical composition between the dendrite and interdendrite regions (Fig. 2) and multiplies the number of sites on the cast surface that promote the ion release process. Alloy "aging" accelerates with increasing number of recastings.

For both elements, a very sensitive correlation between the scale parameter, n, and the recasting number was found (Table III). Specifically a linear correlation was established, $\ln n = a + b/N$, where a is the intercept, and b is the slope. Hence, several decisive conclusions follow: i) the Fe release rate was always higher than that of Co; ii) the release rates increases significantly after two recasting, $i.e.$, eight times and fifteen times faster for Fe and Co, respectively; iii) however, the ion release acceleration decreased with further recasting; iv) the release rates reached their maxima after an infinite number of recasting,; v) then, the release rate of Fe was about twelve times higher than that of Co.

The ion release rates were checked in terms of their temperature dependence. In the interval of the 10th to 22nd week, the temperature of the last aliquots of once and eight times recast samples was below 37 °C, $i.e.$, varied on a daily basis between 18–25 °C. For both elements, a downward deviation of the Weibull linear plots was observed, (Figs. 6 and 7). Obviously, the ion release rate slows down at temperatures lower than those in the human body.

CONCLUSIONS

A commercially available Co–Cr based dental alloy (Dentalite-C) was tested on the effects of multiple recasting. The numbers of recasting were once, four times and eight times. Changes in the structure of the material were followed. In addition, the ion release in artificial saliva was monitored $in vitro$ over 22 weeks at the human body temperature of 37 °C.

The once cast sample had a dendritic solidification microstructure, which was changed by repeated recasting. The changes included microstructure fading, and, consequently, decreases in the macro and micro-hardness, as well as electrical conductivity. The content of the alloy base elements Co and Cr tended to equalize in the dendritic and interdendritic regions.

Only Co and Fe were detected in artificial saliva and their release rates were estimated. The release rate always increased with time regardless of the number of recastings. Multiple recasting substantially accelerated the ion release. How-

ever, the acceleration gradually decreased with increasing number of recastings. After an infinite number of recasting, the release rate reaches a maximum limit value. The release rate of Fe was always higher than that of Co. Nevertheless, the current concentrations of Fe and Co under the given conditions would probably not be considered toxic to humans, because there is no accumulation effect.

The once cast Co–Cr based dental (Dentalite-C) alloy showed durability in the sense of material structure and chemical resistance against artificial saliva and the given conditions.

И З В О Д

НЕКЕ ПОСЛЕДИЦЕ ВИШЕСТРУКОГ ПРЕТАПАЊА ЗУБНЕ ЛЕГУРЕ НА БАЗИ КОБАЛТ–ХРОМ

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У раду је описан утицај вишеструког претапања зубне легуре на бази кобалт–хрома на структурне карактеристике и брзину отпуштања јона у вештачкој пљувачци. Легура је претапана једном, четири и осам пута. Са повећањем броја претапања, микроструктура постаје хомогеније, док се вредности макро и микро тврдоће смањују, као и електрична проводљивост. Отпуштање металних јона у вештачкој пљувачци је праћено "*in vitro*" у периоду од 22 недеље на температури од 37 °C и pH 2,3. У пљувачци су детектовани само јони кобалта и железа и мерене су њихове брзине отпуштања. Утврђено је да је отпуштање јона убрзан процес, односно да брзина отпуштања јона железа и кобалта расте са временом; такође да се брзина отпуштања повећава са повећањем броја претапања, али да се убрзање отпуштања јона смањује са повећањем броја претапања. Брзина отпуштања јона железа је већа него јона кобалта без обзира на број претапања.

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