Long-term corrosion studies in vitro of gold, cobalt–chromium, and nickel–chromium alloys in contact

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Specimens of various types of dental casting alloys were stored in phosphate-buffered 0.9% NaCl solution for 35 weeks. Every 7 weeks the solutions were changed and analyzed with regard to elements released from the alloys. The release of Cu from type III gold alloy increased in contact with gold alloys for metallo-ceramic use in a 1:2 area relation. However, when the area relation was reversed, no difference in the amounts of elements released was observed. Crevice corrosion was initiated with one of the Co–Cr alloys in contact with type III gold alloy. The crevice corrosion increased the amounts of Co and Cr released into the solutions. The release of Ni and Cr from the Ni–Cr alloys was prominent. These alloys were very susceptible to crevice corrosion. With one of the Ni–Cr alloys the release of elements increased in contact with type III gold alloy. ☐ *Copper; crevice corrosion; dental alloys; dissolution; metal release*

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Gold alloys have been ascribed a high degree of corrosion resistance. Nevertheless, copper, silver, and gold from gold alloys have been found in artificial saliva (1). Release of elements from Co–Cr alloys, both in vitro and in vivo, has been reported (2, 3). Ni and Cr from Ni–Cr alloys have also been found in artificial saliva (4).

In vivo penetration of elements into hard and soft tissues of the oral cavity has been reported (5–7). The corrosion products could, in some patients, induce adverse effects such as lesions in the oral mucosa, allergy, and salty or metallic taste (8, 9).

In the mouth, potential differences exist between dental restorations, especially when made of different kinds of alloys (10), and calculations of possible intraoral currents between restorations have been performed (10, 11). In vitro, potentials of dental casting alloys (12, 13) have been measured and currents calculated for 'simulated contact' between different alloys (13).

Contact between restorations made of different kinds of alloys is common in the oral cavity. However, the corrosion effect, with regard to elements released by combining dental casting alloys in contact, has not been reported.

The aim of the present investigation was to study the effect on the corrosion, by measuring the release of metals into a 0.9% NaCl solution, over an extended period of time, when various types of dental casting alloys were in contact.

Materials and methods

The alloys used in the investigation are presented in Tables 1 and 2. The compositions of Co–Cr and Ni–Cr alloys were not declared by the manufacturers, and the surfaces of the castings were therefore analyzed by scanning electron microscopy (EDAX; Philips SEM 501, Philips, Eindhofen, The Netherlands). The concentrations represent an average value, paying no attention to segregations. Constituents with low atomic number, such as boron and carbon, and metals present in concentrations less than 1% by weight were not detected by this method. Six days of immersion in HNO_3 and HCl (1:3; 4 ml) did not dissolve detectable amounts of Be

Table 1. The dental casting gold alloys used and their compositions according to the manufacturers, expressed in percentage by weight. Within parentheses the abbreviations used in the study

Metal	24k gold	Sjödings C* (JSC)	Sjödings M† (JSM)	Matticraft G‡ (MG)	Herador G§ (HG)	Jelenko 'O'¶ (Jel 'O')
Gold	99.9	76.5	87.8	86.0	82.64	87.5
Platinum		2.95	4.3	10.6	16.02	4.5
Palladium			5.8	0.1		6.0
Copper		11.5			0.18	
Silver		8.5	0.8	0.2		1.0
Zinc		0.5				
Iron			0.4			0.25
Indium			0.4	2.0	0.86	0.25
Iridium		0.05	0.1	0.1	0.17	
Tin			0.4			0.5
Rhodium				1.0		

* Conventional type III dental casting gold alloy. Sjödings, Spånga, Sweden.

† Gold alloy for ceramic fused to metal restorations. Sjödings.

‡ Gold alloy for ceramic fused to metal restorations. Johnson Matthey Metals Limited, London, UK.

§ Gold alloy for ceramic fused to metal restorations. Heraeus Edelmetalle GmbH, Hanau, FRG.

¶ Gold alloy for ceramic fused to metal restorations. J. F. Jelenko & Co., Armonk, N.Y., USA.

Table 2. The cobalt–chromium and nickel–chromium dental casting alloys used and their compositions according to SEM (EDAX) analyses made; in percentage by weight. Within parentheses the abbreviations used in the study

Metal	Dentallium* (D)	Biocast† (B)	Wirobond‡ (W)	Ceramalloy§ (C)	Ceramalloy II§ (CII)
Cobalt	68	63	63		
Chromium	19	30	29	17	18
Nickel				72	69
Molybdenum	7	3	7	5	6
Silicon	6	4	1	6	6

* Svedia, Enköping, Sweden.

† Jeneric Gold Co., Wallingford, Conn., USA.

‡ BEGO, Bremer Goldschlägerei Wilhelm Herbst, Bremen, FRG.

§ Ceramco Inc., East Windsor, N.J., USA.

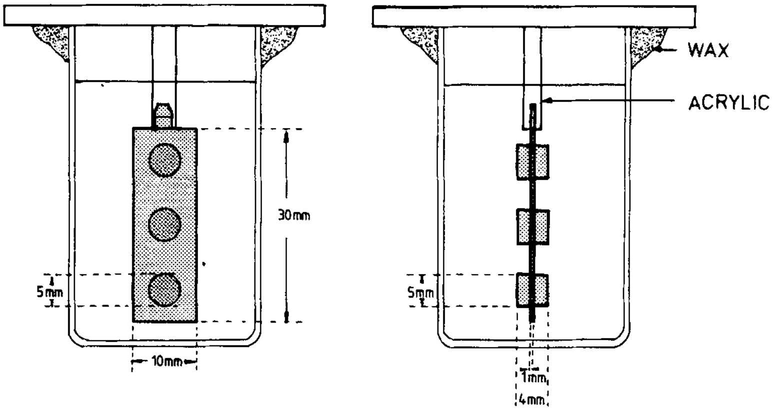

Fig. 1. A plate with three plugs in phosphate-buffered saline solution.

(<0.01 ng/10 mg; atomic absorption spectrophotometry) from samples of the alloys.

The investing, burnout, and pre-heating temperatures and the casting procedures are described in a previous study (2). The specimens were polished down to 600 grit with silicon carbide paper (3M, St. Paul, Minn., USA).

Combinations of plates (6 cm^2) with three plugs (1 cm^2 each), inserted in holes of plates (Fig. 1), were stored in separate beakers with 23 ml 0.9% NaCl solution buffered with phosphate (NaH_2PO_4, 8.8 mM, and Na_2HPO_4, 1.2 mM) to pH 6.0. Five specimens were made of each alloy combination. The beakers were stored at 37 °C and shaken once a day for a few seconds. The experiment was in progress for 35 weeks. Every 7 weeks the solutions were renewed. The specimens were brushed lightly in the solutions, which were analyzed with regard to Cu, Zn, Ag, Pt, Pd, Au, Fe, Co, Cr, Ni, and Mo by means of an atomic absorption flame spectrophotometer. The experimental and analytical procedures have been described previously (2).

The amounts of metals released varied over a wide range, and standard deviations increased approximately proportional to the mean values. To obtain homogeneity of the variances, the amounts measured were transformed into logarithms and tested for by Student's *t* test for two means. A significance level of $P < 0.01$ was used.

Results

Effects on the gold alloys

The release of Cu and Zn from type III gold alloy was greatest the first 7-week period and decreased the second 7-week period (Table 3). After the 3rd or 4th 7-week period, however, the release of Cu and Zn was increased. Ag, Au, and Pt were found in small quantities in some solutions with type III gold alloys, especially in the first 7-week period. No Pd was detected in the solutions. Only small amounts of Zn were found in the solutions when gold alloys for metallo-ceramic use were immersed.

Table 3. The amounts of Cu and Zn released from type III gold alloy in the saline solution during the 35 weeks. Mean (M) and range values of five specimens are presented

	Amounts of metals released (μg/cm^2)			
	Cu		Zn	
Weeks	M	Range	M	Range
0–7	2.2	1.8–2.5	0.8	0.3–1.2
8–14	0.4	0.3–0.5	<0.1	ND*
15–21	1.3	0.5–3.1	0.3	ND–0.8
22–28	0.4	0.3–0.6	<0.1	ND
29–35	0.6	0.4–0.9	<0.1	ND
Total	4.9	4.0–6.8	1.1	0.3–1.5

* ND = not detected.

Table 4. The amounts of Cu and Zn released from type III gold alloy plugs and gold alloys for metallo-ceramic use (plates) in contact in the saline solution for 35 weeks (accumulated values from measurements every 7 weeks) compared with the alloys immersed separately. When immersed in separate solutions, the amounts released from the plugs and the plates were added. Mean (M) and standard deviation (SD) of five combinations are presented

	Amounts of metals released (μg/cm^2)							
	Cu				Zn			
	Separately		In contact		Separately		In contact	
Plates	M	SD	M	SD	M	SD	M	SD
Sjödings M	4.9	(1.1)	10.6*	(3.0)	1.4	(0.5)	3.6	(3.3)
Matticraft G	4.9	(1.1)	9.2*	(1.2)	1.2	(0.4)	1.5	(0.7)
Herador G	4.9	(1.1)	12.5*	(2.9)	1.5	(0.7)	3.5	(1.0)

* Significant change, $P < 0.01$.

Table 5. The amounts of Cu and Zn released from type III gold alloy plates and a gold alloy for metallo-ceramic use and an Ni–Cr alloy (plugs) in contact in the saline solution for 35 weeks (accumulated values from measurements every 7 weeks) compared with the alloys immersed separately. When immersed in separate solutions, the amounts released from the plugs and the plates were added. Mean (M) and standard deviation (SD) of five combinations are presented

	Amounts of metals released ($\mu g/cm^2$)							
	Cu				Zn			
	Separately		In contact		Separately		In contact	
Plugs	M	SD	M	SD	M	SD	M	SD
Sjödings M	4.9	(1.1)	6.4	(1.9)	1.2	(0.3)	3.3	(1.8)
Ceramalloy II	5.8	(1.5)	5.2	(1.1)	1.6	(0.4)	2.1	(0.8)

No significant changes ($P < 0.01$) were found between specimens immersed separately and in contact.

When gold alloys for metallo-ceramic use (plates, 6 cm^2) were placed in contact with type III gold alloy (plugs, 3 cm^2), the amount of Cu released into the solutions increased (Table 4). No such increase was observed with Zn. With the reversed area relation, the amount of elements released did not change when the alloys were placed in contact (Table 5).

Effects on the Co–Cr alloys

The amount of Co released into the solution from the Co–Cr alloys was high the first 7-week period but decreased during the second 7-week period (Table 6). An increase in the amount of Co or Cr released was observed with some specimens the third or fourth 7-week period, especially when placed in contact with the gold alloys. Moreover, Mo, in the range corresponding to 0–5.4 $\mu g/cm^2$ of the plates, was found in solutions with large amounts of Co and Cr.

There was an increase in the amount of Co and Cr released into the solutions from Biocast when stored in contact with the gold alloy (JSC) (Table 7). In contact with the gold alloy the release of Co and Cr from Wirobond was limited to the initial periods, whereas the release of Co from Dentallium

Table 6. The amounts of Co and Cr released from the Co–Cr alloys in the saline solution during the 35 weeks. Mean values (M) and range of five specimens (plates) are presented

	Amounts of metals released ($\mu g/cm^2$)											
	Dentallium				Biocast				Wirobond			
	Co		Cr		Co		Cr		Co		Cr	
Weeks	M	Range	M	Range	M	Range	M	Range	M	Range	M	Range
0–7	4.0	ND*–9.5	3.0	0.7–5.2	2.5	1.4–5.5	<0.6	ND	6.2	2.8–11.5	<0.6	ND–0.6
8–14	0.4	ND–1.5	<0.6	ND–0.6	<0.1	ND	<0.6	ND	0.7	0.1–1.5	<0.6	ND
15–21	0.5	ND–0.9	<0.6	ND	<0.1	ND	<0.6	ND–0.6	0.4	ND–0.9	0.6	ND–1.1
22–28	0.4	ND–0.5	<0.6	ND–0.6	0.6	ND–2.9	<0.6	ND	<0.1	ND	<0.6	ND
29–35	<0.1	ND–0.1	<0.6	ND	<0.1	ND	<0.6	ND	<0.1	ND	<0.6	ND
Total	5.0	ND–12.2	3.9	1.3–5.6	3.1	1.4–8.3	<3.0	ND–0.6	7.3	2.9–13.9	<3.0	ND–1.7

* ND = not detectable.

Table 7. The amounts of Co and Cr released from the Co–Cr alloy plates in contact with type III gold alloy plugs in the saline solution for 35 weeks (accumulated values from measurements every 7 weeks) compared with the alloys immersed separately. Mean (M) and standard deviation (SD) of five combinations are presented

	Amounts of metals released (μg/cm²)							
	Co				Cr			
	Separately		In contact		Separately		In contact	
Co–Cr alloys	M	SD	M	SD	M	SD	M	SD
Dentallium	5.0	(4.6)	14.1	(15.4)	3.9	(1.7)	<3.0	
Biocast	3.1	(3.0)	116*	(103)	<3.0		33*	(36)
Wirobond	7.3	(4.5)	8.3	(10.2)	<3.0		<3.0	

* Significant changes, $P < 0.01$.

and Cr from Biocast occurred in all 35 weeks. The increased average values for Co, when Dentallium and Wirobond were immersed in contact with the gold alloy, were due to the fact that one out of five specimens exhibited a particularly high release of Co.

Green corrosion products were observed with Biocast at the acrylic holder of the plates. When separated from the holder the plates showed a rough surface underneath.

Effects on the Ni–Cr alloys

Table 8 shows the amounts of Ni and Cr released into the solutions from the Ni–Cr alloys. The release of Ni and Cr from one of the Ni–Cr alloys (C) increased markedly when placed in contact with type III gold alloy (JSC). The difference between the two Ni–Cr alloys with regard to corrosion was most pronounced when they were stored in contact with JSC.

The C plates showed heavy crevice corrosion and were dull and covered with green corrosion products on the surface. In contrast, the CII plates were shiny and showed less pronounced crevice corrosion.

The presence of JSC caused an increase in the Cr/Ni ratio, in the solutions, by a factor of 7.4 for C plates and 4.6 for CII plates. When the release of elements was high, the pH of the solutions increased. The highest pH, 6.9, was found the first 7-week period with C in contact with JSC. The increased

Table 8. The amounts of Ni and Cr released from the Ni–Cr alloy plates in contact with type III gold alloy plugs in the saline solution for 35 weeks (accumulated values from measurements every 7 weeks) compared with the alloys immersed separately. Mean (M) and standard deviation (SD) of five combinations are presented

	Amounts of metals released (μg/cm²)							
	Ni				Cr			
	Separately		In contact		Separately		In contact	
Ni–Cr alloys	M	SD	M	SD	M	SD	M	SD
Ceramalloy	625	(325)	2104*	(540)	10.8	(4.4)	268*	(168)
Ceramalloy II	242	(412)	399	(139)	8.2	(16.8)	62	(31)

* Significant changes, $P < 0.01$.

pH of the solutions was correlated to the amount of Ni released (correlation coefficient, 0.76).

The release of Mo also increased from the Ni–Cr alloys when they were placed in contact with the gold alloy. With alloy C, Mo release increased from a mean of 0.4 μg/cm^2 (range, 0–1.8) to 43 μg/cm^2 (range, 19–78) during 35 weeks and with CII from 0.2 μg/cm^2 (0–4.0) to 19 μg/cm^2 (6–31).

Etching of the type III gold alloy after contact with the Co–Cr alloys and the Ni–Cr alloys

No tarnish was observed on the surface of the type III gold alloy after immersion in contact with the Co–Cr or the Ni–Cr alloys.

When the gold alloy plugs were etched in HCl (2.4 M) for 10 min after having been in contact with Ceramalloy for 35 weeks, Ni, 513 ± 106 μg/cm^2 (mean ± SD), and Cr, 7.5 ± 3.0 μg/cm^2, were found in the acid. The corresponding values for gold alloys that had been in contact with Ceramalloy II were lower, 0.3 ± 0.1 and <0.3, respectively. When the gold alloys were immersed in contact with Co–Cr alloys, neither Co, <0.05 μg/cm^2, nor Cr, <0.03 μg/cm^2, was found in the acid after etching.

Discussion

Release of elements from all the casting alloys tested was observed in the saline solution. The release of corrosion products decreased with time. However, elements continued to be released from type III gold alloy and the Ni–Cr alloys throughout the experiment. When one of the Co–Cr alloys (B) and one of the Ni–Cr alloys (C) were in contact with type III gold alloy (JSC), an increase in the amount of elements released was observed. This was also the case with type III gold alloy in contact with all the metallo-ceramic gold alloys at a 1:2 area ratio. However, at an area ratio of 2:1, no increase was registered.

To keep the number of factors low, which could influence the corrosion process, and to make comparisons with previously published data possible, the 0.9% NaCl solution was used to study the galvanic effects. This Cl$^-$ concentration is about six times larger than the Cl$^-$ concentration in the saliva. However, many solutions more aggressive than saliva will sometimes surround dental restorations. In the present study chlorides formed by the elements of the alloys were dissolved in the solution, which explains the absence of tarnish on the surfaces.

Brune et al. (1) have studied the release of Cu, Ag, and Au from gold alloys by means of a nuclear trace technique. The amounts of the various elements found in the solutions fall within the range of those measured in the present study.

The release of Cu from type III gold alloy (3 cm^2) increased in contact with the metallo-ceramic alloy (JSM; 6 cm^2). Gold alloys for metallo-ceramic purposes have been shown to possess a more electro-positive potential than type III gold alloys—that is, their ability to act as a cathode when coupled to another alloy is greater (12). It is also known that an increased cathode to anode relation could make the anodic attack stronger (14). The elements liberated from the alloys will also be dependent on the electrolyte.

The release of Co from the Co–Cr alloys not in contact with JSC was more prominent than that of Cr. The mean amount of Co analyzed in the solutions after 7 weeks in the present study was 4.2 μg/cm^2 (range, <0.4–11.5) ($n = 15$). Release of Cr was observed only for Dentallium, 3.0 μg/cm^2 (0.4–5.2) ($n = 5$), whereas release from Biocast and Wirobond was undetectable (<0.6 μg/cm^2). Espevik (4) found that the concentrations of Co and Cr after 2 months in artificial saliva were below the detection limit (which corresponded to 6.25 μg/cm^2 released from the specimens). Brune et al. (15), after about 7 weeks, found a release of 3.4 μg Co and 0.63 μg Cr/cm^2 from a Co–Cr alloy into artificial saliva.

The experimental design in the present study gave rise to a crevice between the acrylic holder and the plates. When the plates were placed in contact with the gold alloy, the crevice was even more exposed to anodic attack, and the release of corrosion products increased. With Biocast the increase in elements released in the presence of gold probably originated mainly from crevice corrosion. Dentallium and Wirobond showed a more resistant behavior than Biocast. The relatively low concentration of Mo in Biocast, 2.6%, as compared with about 7% for Dentallium and Wirobond, could explain the higher susceptibility of Biocast to corrosion (cf. 16). The release of elements from the Co–Cr alloys decreased with time. A selective release of Co and the formation of a protective film on the surface, containing Cr–O or Cr–OH, have been proposed as explanations for the passivation of Cr–Co–Mo-based alloys (15).

The high susceptibility of Ni–Cr alloys to corrode in crevices is in agreement with earlier investigations (17). The release of elements from one of the alloys, Ceramalloy, increased when stored in contact with the gold alloy. The lower content of Mo, 5.1%, in Ceramalloy as compared with 6.1% in Ceramalloy II, and the difference in microstructure, with more eutectic-like shapes and with more extensive precipitates in Ceramalloy (18), might explain the greater susceptibility of Ceramalloy to corrode.

Ni was more prone to form soluble compounds (hydroxide) in the saline solution than was Cr. When the Ni–Cr alloys were anodes in contact with the gold alloy, the Cr/Ni ratio increased in the solutions, suggesting a breakdown of the passivating layer formed by Cr. The corrosion products that accumulated at the opening of the crevice, probably metal hydroxides (14), were not measured. An earlier study (2) showed that in a saline solution of pH 4 less precipitations were formed on the surface of the specimens. In the oral cavity these precipitations may not form. The release of corrosion products for both the Ni–Cr alloys continued the entire 35-week period without any tendency to decrease.

The increase in corrosion products found in the solutions after 21 or 28 weeks was observed for all the alloys. A delayed release of elements after total hip replacements (19) and insertion of dental Co–Cr constructions (3) has been observed earlier. This delay may partly be due to polishing procedures used in technical and clinical practice, which might form a film on the surface of the alloys, inhibiting corrosion in the initial period (20).

All samples examined in the present study were in 'as-cast' condition. However, some of the alloys, before they are used clinically, receive additional thermal treatments, for instance during baking on porcelain. However, no change in the corrosion of Co–Cr and Ni–Cr alloys due to heat treatment has been reported (21, 22).

Although there exist investigations on the possible effect of other ions and proteins on the corrosion process, an isotonic saline electrolyte has often been utilized for electrochemical corrosion experiments. Revie & Greene (23) have demonstrated in dog and rabbit models that the corrosion rates of stainless steel with isotonic saline solution in vitro are comparble to or less than the corrosion rates measured in vivo. Furthermore, Buchanan & Lemons (24) have shown a close correlation between in vitro polarization results of Co–Cr alloys in isotonic saline solution and in vivo polarization in rabbits. Hensten-Pettersen & Jacobsen (25) reported reduced dissolution of elements from dental base-metal alloys in the presence of calf serum.

The amount of Cu released within the initial 3 weeks from type III gold alloy in the present study (approximately 1 $\mu g/cm^2$) corresponds well with the amount measured in dentin under a type III gold full crown in the oral cavity over the same period (7). The release of Co and Cr from a circumferential clasp with an occlusal rest has been measured in vivo in the tooth substance after 3 weeks in contact (7). The amount found in that tooth substance could be extrapolated to about 0.2 µg Co and 0.4 µg Cr. Considering the fact that Co and Cr were also released into other tissues and fluids, the total amount released in vivo and in vitro in this study seems to be of the same magnitude.

The oral environment is complex with many aggressive or inhibiting factors active on the corrosion process. Crevice corrosion is, to a great extent, self-propagating and less influenced by the surrounding electrolyte solution. Therefore, placing different kinds of dental casting alloys in contact with gold could be a potential risk for increased amounts of elements being released from these alloys.

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