From: The Odontological Faculties of Karolinska Institutet, Stockholm and of the University of Umeå, Umeå, Sweden.

STUDIES OF THE TRANSPORT OF METAL IONS FROM GOLD INLAYS INTO ENVIRONMENTAL TISSUES

by

Axel Bergenholtz

Björn Hedegård

Rune Söremark

The concentration of some micro-elements in "normal" human enamel and dentine has been the subject of investigation (*Samsahl & Söremark,* 1962). Using this study as a background, further investigation was performed concerning the influence of prosthetic applications on the concentrations of micro-elements in human teeth (*Söremark et al.,* 1962). The teeth were analysed after having been restored with gold inlays, silver amalgam, and gold crowns, and after having had gold and chromium-cobalt alloy clasps in contact with their enamel surfaces for given periods of time. These experiments showed that elements present in the various restorative materials penetrated to a large extent into the hard tissues of the teeth.

The role of galvanic action due to the potential difference between the metals of the reconstructions, the saliva serving as an electrolyte, has often been discussed (*Ullmann,* 1932; *Lain & Caughron,* 1936; *Spreng,* 1940; *Rheinwald,* 1953). Some soft tissue irritations, sometimes in the form of ulcerations, sometimes as epithelial changes of various intensity of hyperkeratosis are supposed to be caused by the galvanic action. However, there is also reason to suspect some form of metalosis, set up by the deposition of metal ions in the soft tissues.

Ion transport, presumably due to galvanic action, seems to be more pronounced with untreated "non-homogenized" than with heat-treated "homogenized" gold alloys (*Hedegård,* 1958).

Since, however, all these studies have been performed on persons with tooth restorations of dissimilar alloys, they do not enable the basic pattern of the metal ion transport to be ascertained; further experimental work was considered necessary to resolve this problem.

The purpose of the present investigation was:

a) to compare the penetration of metal ions into the dental hard tissues with that into the gingival tissues and,
b) to examine the possible difference in the transport into enamel and dentine and into environmental gingiva of metal ions from various types of gold alloys and from heat-treated ("homogenized") and untreated cast gold inlays.

MATERIAL AND METHODS

This study had to be performed on experimental animals. Three 1½ years old sibling dogs in general good health were used for the experiments. Class V inlay cavities were prepared in premolars, canines, and incisors. In all, 42 teeth were prepared. Gold inlays were prepared according to ordinary technique. Twenty-carat gold (supplied by Ädelmetallbolaget), C- and E-gold alloys (supplied by Sjödings and Co.) were used for the inlays. All the inlays were boiled in acid, rinsed in water, dried and fixed in the cavities with Phosphatine (a phosphate cement). During the time interval between the preparation of the cavities and the cementing of the inlays, the cavities were sealed with temporary stopping.

Half the number of the inlays of each gold type were heat-treated ("homogenized") at 650° C for 1½ hours. On each dog, cavities were prepared on one side of the upper and lower jaws, inlays fitted and cemented and left for 15 days. After extraction of these teeth for analyzing and a subsequent rest period, gold inlays of the same type of gold but with different heat-treatment were fitted to teeth in the other side of the mouth and left for an equal period of time, 15 days.

All samples were dried in an electric oven for 48 hours at 65° C. The dried samples were then placed in polyethylene bags and sealed. The samples were not in contact with materials other than glass or polyethylene. Standard amounts of the three elements to be studied, Au, Zn, and Cu, were inserted into polyethylene tubes and sealed and then placed in the same standard aluminium can as the sample. Thus, the standards and the samples were irradiated simultaneously. The aluminium can containing the standards and the samples to be studied was irradiated by thermal neutrons for about 20 hours. The flux was about 10^{12} neutrons $\times$ cm^{-2} $\times$ sec.$^{-1}$. When the period of irradiation was completed, the tooth samples were dissolved in 1 N HCl containing 0.3 % H_2O_2, and the gingival samples were dissolved in H_2SO_4. The group separation technique by *Samsahl & Söremark* (1962) was used where applicable. The gamma-ray spectra were then registered in a single channel analyzer operating with a well-type scintillation crystal (3″ $\times$ 3″).

The entire tooth was used, the enamel and dentine thus not being separated. Records of one tooth sample and one gingiva sample are given in Figs. 1 and 2.

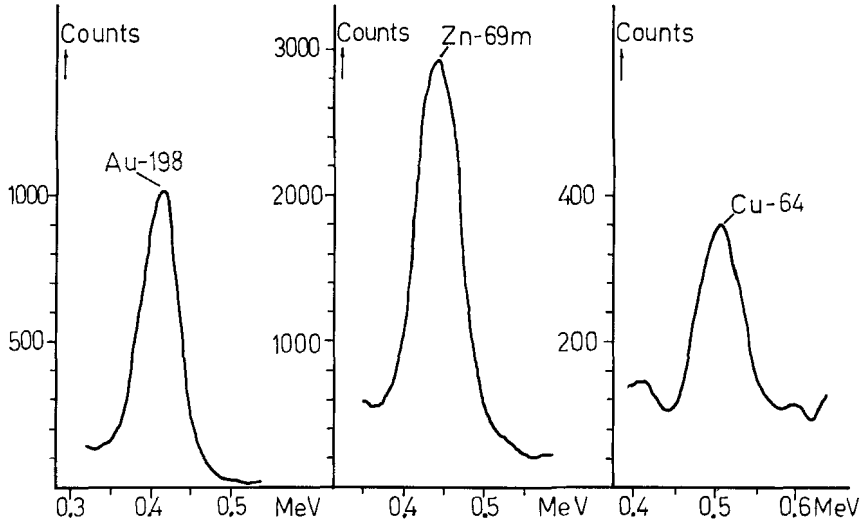

Fig. 1. Records of one tooth sample (heat-treated gold inlay).

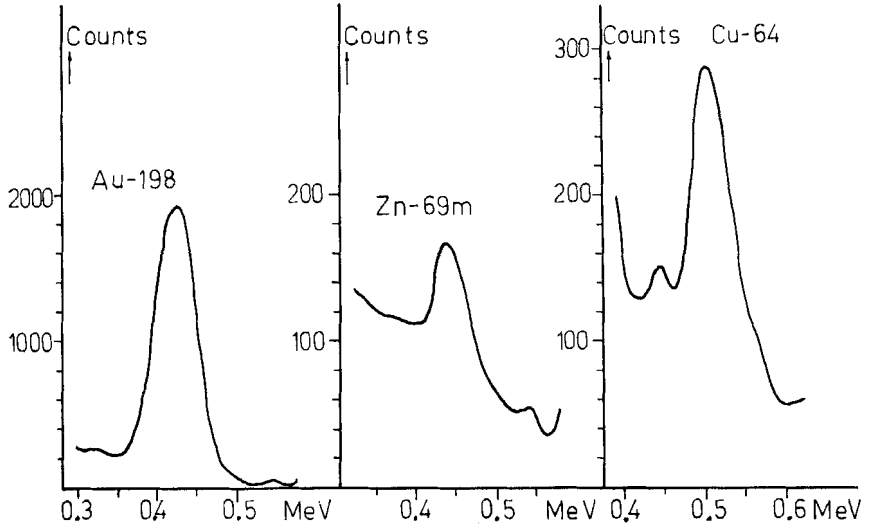

Fig. 2. Records of one gingiva sample (heat-treated gold inlay).

Reproducibility tests of the analyzing method used

To ascertain the reproducibility of the method, the following tests were made. After neutron irradiation of three dog tooth samples (about 200 mg each) at different times for about 20 hours in the flux of about 10^{12} neutrons $\times$ cm^{-2} $\times$ $sec.^{-1}$, the samples were dissolved as described. Of the solution obtained (50 ml), 4 portions were taken, each of which was radiochemically separated in groups. Two laboratory assistants each analyzed independently 2 portions of the concentrations of gold and zinc. In this way three different tooth samples (I, II, III) were tested. To one of the samples (III) 1 ml of a gold and zinc solution was added as carrier before dissolving. The results of the reproducibility tests are given in Table I.

The present tests of the reproducibility showed a close agreement among the different portions from the same tooth sample. In most cases the differences between the various portions from the same tooth sample were about the same as the standard deviation of the impulse counting. The results obtained when carrier was added before the chemical group separation (sample III) were not of higher reproducibility.

Table I

Results of the reproducibility tests concerning the analysis of the concentrations of gold and zinc in dog teeth

	Sample	Au^{198}	Zn^{69m}
Counting rate, mean of four portions and the standard deviation in %	I	306 ± 8.1	2140 ± 4.7
	II	182 ± 9.4	1620 ± 9.9
	III	133 ± 24.0	1560 ± 9.0
The standard deviation of the counting rate in %	I	6.9	2.2
	II	7.4	2.5
	III	1.8	2.5
The deviation between the two laboratory assistants in %	I	0.5	4.6
	II	0.3	5.7
	III	1.1	4.6

No carriers were used in the radiochemical separation procedures in the studies presented in the next section. The method of analysis was considered to be reliable for the present study.

RESULTS

Tables II and III present the amount of Au, Zn, and Cu found in the teeth and in the marginal gingiva after a period of fifteen days with the inlays of various gold alloys *in situ*. Table IV gives the test values for the controls.

The results showed high concentrations of Au, Zn, and Cu in the hard tissues of all the teeth with gold inlays and in the marginal gingiva contacting these teeth. The concentrations showed, however, a considerable fluctuation.

The results did not show any correlation with various types of gold alloys tested nor were there any significant differences associated with the heat-treatment of the inlays.

It was not always possible to obtain data on all three metals for each specimen due to difficulties in organizing the measuring operation in the short time available for the two elements with fast decreasing activity (Cu and Zn).

Table II

Amounts (ppm) of Au, Zn, and Cu in dog teeth with gold inlays *in situ* for fifteen days

Dog I

20 carat gold alloy,

heat-treated (homogenized)				not heat-treated (non-homogenized)			
Tooth	Au	Zn	Cu	Tooth	Au	Zn	Cu
—P1	0.36	508	6.59	P1—	6.33	4250	13.5
—P2	9.81	428	59.6	P2—	0.93	528	1.63
—P3		301	17.8	P3—	3.06	472	17.2
				P4—	0.055	12.5	
+P2	0.93	1610	5.8	P2+	0.40	815	3.08
+P3	32.8	331	1.98	P3+	10.56	495	3.03
+P4	21.5	378	15.65	P4+	8.35	1785	11.3

Dog I and II

C-gold alloy,

heat-treated (homogenized)				not heat-treated (non-homogenized)			
Tooth	Au	Zn	Cu	Tooth	Au	Zn	Cu
—P1	1.7	9763	0.9	P1—	2.6	531	1.7
—P2	7.9	14647	3.4	P2—	60.6	5430	16.6
—P3	3.5	5204					
+P2	5.8	2207	2.0	P2+	10.5	945	1.7
—I3	40.0	5380					
+I2	0.20	8308	4.9	I2+	11.1	2870	2.1
+I3	0.25	99					
—C1	1.5	903		C1—	3.1		18.5
+C1	0.42	1512	1.3	C1+	0.065	68.5	

Dog III

E-gold alloy,

heat-treated (homogenized)				not heat-treated (non-homogenized)			
Tooth	Au	Zn	Cu	Tooth	Au	Zn	Cu
—I3	0.063	234		I3—	0.16	3025	2.3
+I2	1.3	17693	46	I2+	4.7	395	1.6
+I3	0.1	862	49	I3+	0.45	915	0.78
—C1	0.23	3684		C1—	8.5	984	0.88
+C1	0.13	281		C1+	0.098	106	0.82

Table III

Amounts (ppm) of Au, Zn, and Cu in marginal gingival tissue in contact with buccal gold inlays *in situ* for fifteen days in dog teeth

Dog I
20 carat gold alloy,

heat-treated (homogenized)				not heat-treated (non-homogenized)			
Tooth	Au	Zn	Cu	Tooth	Au	Zn	Cu
—P1	0.89			P1—	11.0	346	28.4
—P2	0.82		30.1	P2—	41.6	370	7.4
—P3	51.8		4.6	P3—	396.5	880	112
+P2	1.28	505	22.5	P2+	78.34	252	24.7
				P3+	4.22		13.75
				P4+	42.0		32.5

Dog I and II
C-gold alloy,

heat-treated (homogenized)				not heat-treated (non-homogenized)			
Tooth	Au	Zn	Cu	Tooth	Au	Zn	Cu
—P1	5.1	304	5	P1—	0.81	410	113
—P2	0.27	231	4.6				
—P3	11.2	1481	13.9				
+P2		227	2.2				
—I3	209	230					
+I2	0.2	148	11.8	I2+	384	857	6.9
+I3	1.9	386	11.3	I3+	0.48	248	11.8
—C1	55	355	6.9	C1—	3.4		8.9
+C1	2	1.7	8.9	C1+	0.9	102	11.3

Dog III
E-gold alloy,

heat-treated (homogenized)				not heat-treated (non-homogenized)			
Tooth	Au	Zn	Cu	Tooth	Au	Zn	Cu
—I3	71.4	3885	99	I3—	90.4	102	11.6
+I2	27.4	873	29.5	I2+	0.19	102	1.5
+I3	4.9			I3+	6.6	183.2	8.7
—C1		557	3.3	C1—	0.89	59	3.3
+C1		521	70	C1+	0.11	49.2	2.2

DISCUSSION

As in the previous study by *Söremark et al.*, 1962, elements present in the gold inlays penetrated to a great extent into environmental tissues and increased the concentrations therein.

As the inlays were *in situ* for only one period it was not possible to determine the rate of transport of the metal ions from the restorations into the teeth and the marginal gingiva. However, in the earlier study, it was concluded that the transport was of long duration. The present findings also suggest that the migration continues over a long period.

It is possible that the rate of transport differed from one tooth

Table IV

Values for controls (ppm)

Dog and tooth	Values for the teeth		
	Au	Zn	Cu
Dog I —P4	0.0095	204	0.36
I M3—	0.0145	281	
II M3—	0.0406	204	3.7
III M3—	0.0071	298	
III —I2	0.0014	312	
III —I1	0.055	124	
M ± S. D.	0.021 ± 0.019	237 ± 66	

Dog and tooth	Values for the gingiva		
	Au	Zn	Cu
Dog I —P4	0.68	291	7.0
I C1+	0.098		4.16
I +C1	0.25	96	17.5
I M1+	0.173	240	5.5
II C1+		250	2.9
III P1+	0.0332	246	6.3
III —P2	0.14		
M ± S. D.	0.27 ± 0.21	224 ± 66	7.2 ± 4.7

to another for the concentrations varied widely although the period of exposure was the same throughout (15 days); but this fact may also be due to possible non-controllable variables. Since the size of the gold inlay was limited by the size of the individual tooth the inlays vary considerably in this respect. The area of contact between the gold surface and the tissues was not therefore equal in all the cases. Furthermore, there was no way of preventing gingival retraction. Pathological changes in the marginal gingiva due to smears, mechanical trauma caused through preparation of the cavity and the temporary filling may vary considerably from tooth to tooth and influence the uptake of metal ions. Since, then uncontrollable factors may have an important effect on the rate of metal transport; and, therefore, the results will not be precise.

The comparatively small variation in concentration of the metals tested in the controls suggests that the test procedure is reliable. As the controls were taken on the same occasions as the test samples and subjected to the same treatment, there is no reason to suspect any uncontrolled factor in the test procedures.

It has already been shown by *Söremark et al.* (1962), that single constructions attached to a tooth will deliver metal ions to the dental hard structures. The present study confirms this and also shows that the same is true of the gingiva.

In most cases the concentration of gold and copper was higher in the marginal gingiva than in the tooth tissue, while the reverse was true for zinc. Consequently, the Au and Cu transport into the teeth was slower than that into the soft tissues, which could be expected (*cf.* the review by *Brudevold & Söremark*, 1964). This might be attributed to a retarding effect on the ion transport into the enamel and dentine by the zinc phosphate cement. The cement may partly explain the high concentration of zinc in the tooth tissue.

The values obtained for the "normal" concentration of gold and zinc in the control teeth were found to be in agreement with those obtained for human teeth (*Samsahl & Söremark*, 1962). Too few test results were obtained to allow comparison for copper.

SUMMARY

The penetration of gold, zinc, and copper ions from gold inlays into tooth tissues and adjacent marginal gingiva was studied by means of thermal neutron activation analysis in combination with gamma spectrometry. Three adult dogs were used for the experiment. Forty-two tooth samples and forty-two adjacent gingival samples were analyzed after the gold inlays had been *in situ* for a period of fifteen days. The results were compared to control samples.

The results showed that Au, Zn, and Cu from gold inlays penetrated to a great extent into environmental hard and soft tissues.

The penetration of gold and copper into the marginal gingiva was more pronounced than that into the tooth substances. The reverse was true for zinc.

The present study did not show any differences in the ability of metal ions from different gold alloys to penetrate into the surrounding tissues. C-, E-, and 20-carat gold alloys and heat-treated ("homogenized") and untreated cast gold inlays had the same effect.

RÉSUMÉ

ÉTUDES SUR LE TRANSPORT DANS LES TISSUS ENVIRONNANTS D'IONS MÉTALLIQUES PROVENANT D'INLAYS D'OR

La pénétration dans les tissus dentaires et dans la gencive marginale avoisinante d'ions d'or, de zinc et de cuivre provenant d'inlays d'or a été étudiée par analyse par activation thermique neutronique combinée à la spectrométrie par rayons gamma. Trois chiens adultes ont servi pour cette expérience. Quarante-deux échantillons de dent et quarante-deux échantillons gingivaux adjacents ont été analysés après que les inlays d'or aient été *in situ* pendant une période de quinze jours. Les résultats ont été comparés à ceux obtenus sur des échantillons témoins.

Les résultats ont montré que Au, Zn, et Cu provenant des inlays d'or pénétraient considérablement dans les tissus durs et dans les tissus mous environnants.

La pénétration de l'or et du cuivre dans la gencive marginale

était plus marquée que dans les tissus dentaires. L'inverse était vrai pour le zinc.

La présente étude n'a pas montré de différence entre l'aptitude des ions métalliques provenant de différents alliages d'or à pénétrer dans les tissus environnants. Des alliages d'or de type C, de type E et à 20 carats, et des inlays d'or coulé traités par la chaleur ("homogénéisés") et non traités avaient la même action.

ZUSAMMENFASSUNG

UBER DAS EINDRINGUNGSVERMÖGEN DER METALLIONEN VON GOLD-INLAYS IN DIE UMGEBENDEN GRWEBE

Das Eindringungsvermögen der Gold-, Zink- und Kupferionen von Goldinlays in Zahngewebe und umgebendes Zahnfleisch wurde durch Aktivierungsanalyse sowie mit Gamma Spektrometrie studiert. Drei erwachsene Hunde wurden für den Versuch verwendet.

42 (zweiundvierzig) Zahnproben und 42 Zahnfleischproben wurden analysiert, nachdem die Goldinlays fünfzehn Tage lang in situ gewesen waren. Die Resultate wurden mit Kontrollproben verglichen.

Die Resultate zeigten, dass Gold, Zink und Kupfer der Goldinlays in die umgebenden harten und weichen Gewebe im beträchtlichen Umfang eindrangen.

Das Eindringungsvermögen von Gold und Kupfer in das Zahnfleisch war deutlicher als das in die Zahnsubstanzen. Das Gegensatz galt für Zink.

Diese Arbeit zeigte keine Unterschiede betreffs des Vermögens der Metallionen der verschiedenen Goldalloys in die umgebenden Gewebe einzudringen. C-, E-, und 20 Karat Goldalloys — homogenizierte und nicht homogenizierte — hatten dieselbe Wirkung.

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Addresses:

Axel Bergenholtz
Department of Parodontology
University of Umeå
Umeå 2
Sweden

Björn Hedegård
Department of Prosthetics
Karolinska institutet
Stockholm
Sweden

Rune Söremark
Department of Prosthetics
University of Umeå
Umeå 2
Sweden