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Marginal seal composition in amalgam restored teeth of varying marginal leakage

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SUMMARY

An *in vitro* study was performed to ascertain the marginal seal elemental composition of amalgam restored extracted teeth of known marginal leakage. Occlusal cavities were cut in 400 caries free extracted teeth and left unlined or lined with one of 5 bases. A varnish was applied to half the cavities followed by restoration with a low copper or high copper amalgam to produce 20 restoration combinations. The teeth were stored in 1 per cent NaCl and thymol for 3 and 12 months at 20°C whereafter a standard fluorescent dye marginal leakage test was performed on 320 specimens (eight teeth per treatment). Restoration combinations were grouped into 4 seal classes depending on percentage marginal seal achieved for each combination: 0-25 per cent; 26-50 per cent; 51-75 per cent and 76-100 per cent. The 80 remaining teeth (2 teeth per treatment) were fractured to expose the restoration and cavity surface, covered with marginal seal material and this was analysed by energy dispersive X-ray analysis. Elemental seal composition was compared to percentage marginal seal achieved using ANOVA and Tukey's test with significance set at $p<0,05$. Numbers of elemental analysis specimens falling into each marginal seal class was 0-25 per cent=48; 26-50 per cent=18; 51-75 per cent=10; 76-100 per cent=4. Of the 16 elements detected, nine were significantly linked to sealing/leakage: Ca, Cl, Cu, Mg, Hg, P, Ag, Sn and Zn. The findings have a bearing on the improved longevity of amalgam restorations.

OPSOMMING

'n *In vitro* studie is uitgevoer om die elementale marginale seëlsamestelling van amalgaamherstelde verwyderde tande met bekende marginale lekkasie te bepaal. Okklusale kavieteite is voorberei in 400 kariesvrye verwyderde tande en is of sonder voering gelaat, of uitgevoer met een van vyf basisse. Vernis is aan die helfte van die kaviteite gewend waarna herstellings met hoë- of lae koperamalgaam geplaas is in 20 van bogenoemde verskillende kombinasies. Die tande is gestoor in 1 persent NaCl en timol vir 3 en 12 maande teen 20°C waarna 'n standaard fluoresserende kleurstof marginale lekkasietoets uitgevoer is op 320 monsters (agt tande per behandling). Herstellingskombinasies is ingedeel in vier seëlklasse volgens die persentasie seël wat bereik is in elke kombinasie: 0-25 persent; 26-50 persent; 51-75 persent en 76-100 persent. Die oorblywende 80 tande (twee tande per behandeling) is gefraktuur om die aansluiting tussen kaviteit en die herstelling met die marginale seëlmateriaal wat dit bedek, bloot te lê sodat energieverspreidende X-straal analise gedoen kon word. Elementale seëlsamestelling is vergelyk met die persentasie marginale seël wat bereik is met behulp van die ANOVA en Tukey toetse met betekenisvolheid gestel op $p<0,05$. Die aantal elementale analisemonsters wat in elke marginale seëlkas geval het was 0-25 persent (48); 26-50 persent (18); in 51-75 persent (10) en 76-100 persent (4). Van die 16 elemente waargeneem was nege betekenisvol geassosieer met seëling/lekkasie naamlik Ca, Cl, Cu, Mg, Hg, P, Ag, Sn en Zn. Hierdie bevindinge hou verband met verhoogde lewensduurte van amalgaamherstellings.

INTRODUCTION

A gap forms at the amalgam-tooth interface of freshly placed amalgams due to setting contraction. If accompanied by poor restoration technique this gap can reach a width of 600µm before being detected (Söderholm, Antonson and Fischlschweiger, 1989). Fluid flow, continuous with that occurring on the outer restored surface of the tooth, percolates between the cavity wall and the inner wall of the restoration in a process known as marginal leakage. Within the tooth-restoration gap dynamic forces of chemical, mechanical, electrical, thermal and biological origins induce changes in the surface of the restoration and cavity wall, be they in shape, composition or structure. One of the consequences of this amalgam restoration — oral environment interaction impacts on marginal leakage. A deposit is formed within this interface partly as a result of corrosion and closes the gap preventing further ingress of fluids. The formation of this deposit or marginal seal is a complex process involving the various materials used to prepare and restore the cavity, oral physiological factors, the diet and microbiology of the mouth along with their interactions (Jodaikin, 1981). With time, the level of marginal leakage decreases as a result of ongoing seal buildup and ultimately stops when a closed, sealed cavity is achieved.

In order to gain a better understanding of the mechanisms of seal formation, the elemental nature of the marginal seal at

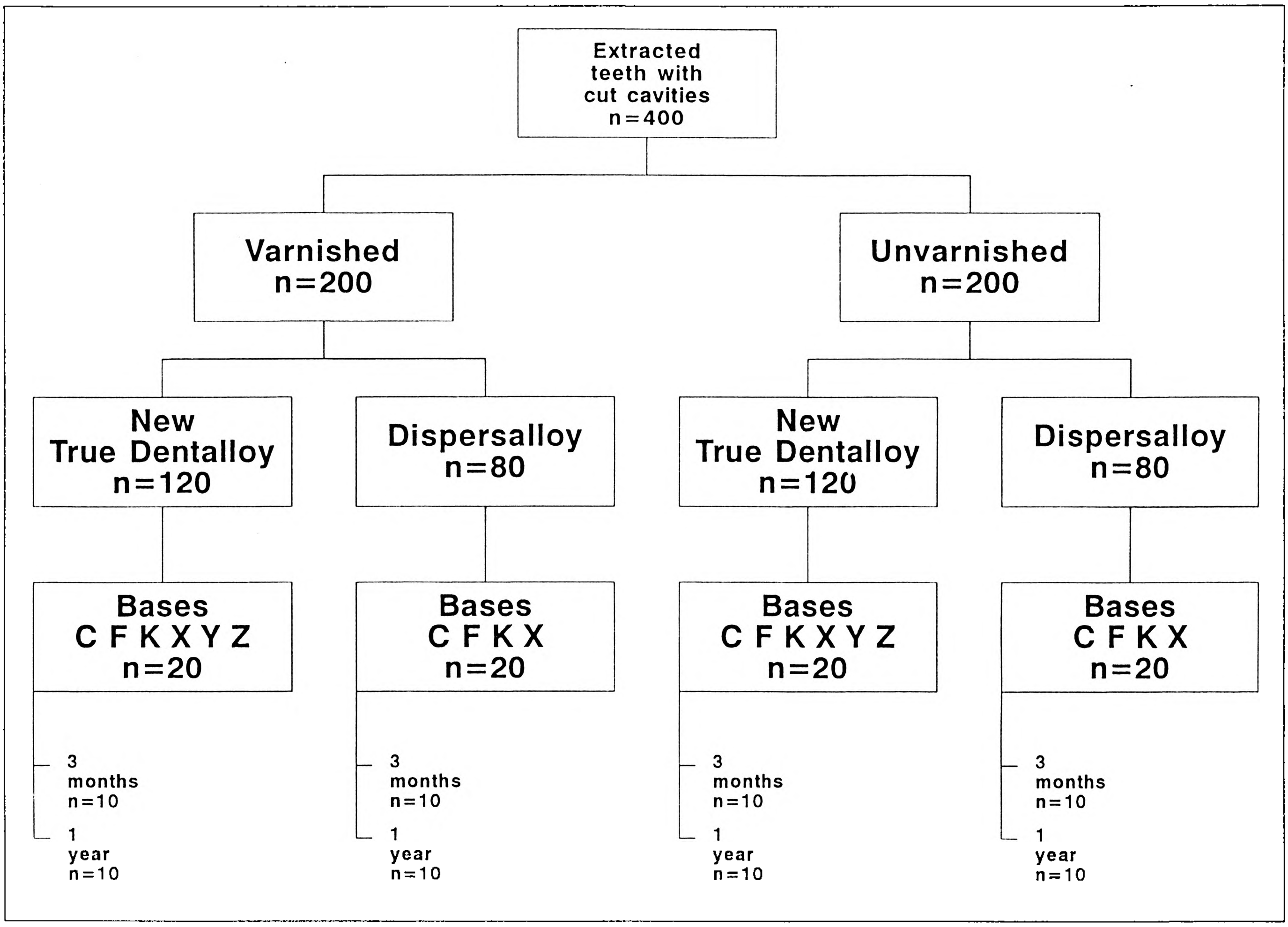

Fig. 1: **Schematic representation of restoration combinations, specimen numbers and ageing times for the specimen group. C=calcium hydroxide, F= Poly-F Plus, K=Kalzinol, X=no base, Y=Dycal, Z=zinc oxide.**

amalgam-tooth interfaces has been examined. Previous studies have generally been retrospective with analyses undertaken on aged, extracted, non-carious amalgam restored teeth where the restoration history was unknown (Sarkar *et al.*, 1975; Grossman, Witcomb and Jodaikin, 1986). Where details of the restorations were available, the degree of cavity sealing has never been established (Marshall and Marshall, 1980; Sarkar *et al.*, 1981; McTigue *et al.*, 1984). Consequently, conclusions drawn from the results of such studies have been applied to marginal seal formation by conjecture without proof that the cavity had actually sealed.

The present study was undertaken to examine the elemental composition of marginal seal material in *in vitro* aged amalgam restored teeth for which the degree of marginal leakage was known.

MATERIALS AND METHODS

The sample consisted of tooth specimens obtained from a pool of restored teeth that were evaluated, in part, for previously reported marginal leakage (Grossman and Matejka, 1993) and marginal seal composition (Grossman, Witcomb and Matejka, 1995) studies. The procedure is summarized briefly. Classical Black's Class I cavities were prepared in 400 extracted, non-carious, human premolars. The teeth were randomly assigned to one of 20 groups of material combinations and restored (Fig. 1). All dental materials selected are regularly used in clinical practice and are listed in Table I. Mixing and insertion of restorative materials were according to manufacturers' instructions. For the two experimental bases zinc oxide and calcium hydroxide, a thick paste of zinc oxide was made by mixing Kalzinol powder with distilled water. Similarly, the calcium hydroxide powder was mixed with distilled water. When used, two coats of varnish were applied to the cavity walls and floor. The teeth were aged in 1 per cent NaCl solution in distilled water with thymol at 20°C for three months and one year. Ten restored teeth were allocated to each time interval per experimental combination.

After each ageing period, eight teeth from each group were subjected to a classical marginal leakage test using 2 per cent Rhodamin B fluorescent dye (E Merck, Darmstadt, Germany). Two transverse, parallel sections were cut at approximately 150-200µm intervals at the middle and outer third of the restorations and examined by transmitted ultraviolet microscopy (Univar, Reichert, Vienna, Austria) to assess the degree of marginal leakage. Sealing was considered to have taken place if penetra-

Table I: *Details of the dental materials used in this study.*

Material	Manufacturer	Batch number	Abbreviation
Polyvar	The Lorvic Corp. 8810 Frost Ave, St Louis, MO 63134 USA	BO32	Varnish
Calcium hydroxide	E Merck. Darmstadt, West Germany.	431A895847	Calc
Poly-F Plus	De Trey, Zurich, Switzerland.	FB61 86/02	PolyF
Kalzinol	De Trey, Zurich, Switzerland/ER Bernard. PO Box 10266, Johannesburg, 2000 South Africa	DC1	Kalz
Dycal	The LD Caulk Division, Dentsply International Inc., Milford, DE 19963-0359 USA	102488	Dycal
Dispersalloy	Johnson and Johnson Dental Products Company, East Windsor, New York, USA	8C816	Cu(high)
New True Dentalloy	S.S. White Manufacturing Ltd. Gloucester, England	033081 # 6798101 and 102684 # 0428410	Cu (low)

tion of dye was up to but not beyond the dentino-enamel junction. Presence of dye beyond the dentino-enamel junction indicated leakage. Percentage seal was calculated as the percentage sections showing sealing for each treatment whereafter the seal scores were allocated to four classes namely 0-25 per cent, 26-50 per cent, 51-75 per cent and 76-100 per cent, for convenience referred to as 25 per cent, 50 per cent, 75 per cent and 100 per cent respectively. Specimens were grouped into these classes irrespective of restorative treatment.

The remaining two specimens from each treatment group of ten were used to determine the elemental composition of the seal material. The restored specimens were fractured to release the marginal seal covered amalgam restoration and expose the marginal seal covered cavity walls. After mounting onto scanning electron microscope specimen stubs these samples were coated with 25nm of spectrographically pure carbon and studied in a JSM-840 scanning electron microscope (JEOL Ltd., Tokyo, Japan) equipped with a LINK AN10000 energy dispersive X-ray analyser (Oxford Instruments Microanalysis Group, Bucks, UK). The latter enabled quantitative area analysis of all elements above atomic number 10 (neon). The analyses were undertaken in two regions on each amalgam specimen, one in the region opposite to the enamel and the second in the area opposite to the dentine. Similarly, area analysis was undertaken on the enamel and dentine of the two separate portions of cut tooth cavity. The area encompassed in each analysis was approximately 0,42mm^2. For the purposes of this investigation the elements analysed in the enamel and dentine areas were pooled. Reference standards of the cavity surface and amalgams were prepared and analysed as above. Between every two analyses a reference spectrum was recorded from a cobalt standard mounted within the specimen stub holder in order to calibrate the energy scale.

Semi-quantitative data of the weight percentage elemental composition of the seal material was obtained by utilizing the peak to background ratios for each element (LINK ZAF-PB/FLS). Analysis conditions were set at an operating voltage of 20kV, an output count rate of 2800 counts/sec and a live counting time of 100 sec.

Use was made of the prepared dental material standard samples to determine the extent of marginal seal deposition on the relevant amalgam and tooth surfaces. Where marginal seal material lying on the amalgam restoration was analysed, comparison of the amount of Ag or Hg detected to the standard can give an indication of seal layer thickness (Anderson, 1973, Grossman *et al.*, 1995). This observation has been extended to the tooth sample by comparing Ca or P present in the analysis of the tooth standard with seal material analysed on the tooth surface. Although such results obtained from contoured specimens are not accurate they do allow comparison on the basis of "thicker" and "thinner".

The data were entered into an IBM 3081-K32 computer (International Business Machines, NY, USA) using SAS (1990) and statistically examined with one way ANOVA and Tukey's studentized range test using the General Linear Models procedure. The critical level of statistical significance chosen was $p<0,05$. Percentage elemental seal composition was compared with the class of seal achieved separately for each time interval and then with both times combined for both the amalgam and tooth analyses.

RESULTS

Marginal leakage

Percentage seal obtained in each specimen combination can be found in Table II. Although sealing improved with time, leakage remained high as more than half the restoration combinations could not attain a sealing of greater than 25 per cent. Restorative materials which were associated with improved sealing were a low copper amalgam, the absence of cavity varnish and a calcium hydroxide paste base. The three month and one year varnish+Calc+Cu(low) and one year varnish+Calc+Cu(high), varnish+Cu(high) and Calc+Cu(high) combinations attained a 75 per cent seal. The combined 100 per cent seal group consisted of the three month and one year Calc+Cu(low) specimens. A more detailed analysis of the marginal leakage results are presented in a previous paper (Grossman and Matejka, 1993).

Elemental analysis

The number of marginal seal analysis specimens destined for each sealing category is listed in Table III. A total of 16 elements were detected in the dental materials used and the marginal seal material of this study: Al, Ca, Cl, Cu, Fe, Mg, Na, Hg, P, Ag, S, Si, Sn, Ti, W and Zn. Of the above elements, nine showed significance when they were linked to sealing:

Table II: *Percentage seal obtained in restoration combinations at both time intervals.*

Restoration combination	Percentage seal 3 months	 1 year
Varnish+Kalz+Cu(low)	6	18
Varnish+ZnO+Cu(low)	6	12
Varnish+Kalz+Cu(high)	0	0
Varnish+PolyF+Cu(low)	18	31
Varnish+PolyF+Cu(high)	6	0
Varnish+Calc+Cu(low)	62	68
Varnish+Dycal+Cu(low)	31	12
Varnish+Calc+Cu(high)	18	56
Varnish+Cu(low)	6	31
Varnish+Cu(high)	6	56
Kalz+Cu(low)	12	37
ZnO+Cu(low)	43	6
Kalz+Cu(high)	6	0
PolyF+Cu(low)	0	25
PolyF+Cu(high)	6	0
Calc+Cu(low)	87	100
Dycal+Cu(low)	18	25
Calc+Cu(high)	43	68
Cu(low)	31	43
Cu(high)	25	37
n	8	8

Where: Varnish=Polyvar; Kalz=Kalzinol; ZnO=Zinc oxide mixed with distilled water; PolyF=Poly-F Plus; Calc=Calcium hydroxide mixed with distilled water; Dycal=Dycal; Cu(low)=New True Dentalloy; Cu(high)=Dispersalloy.

Table III: *Number of tooth specimens subjected to marginal seal analysis in each seal group.*

Seal group	Three months	Specimen no. One year	Combined
25%	28	20	48
50%	8	10	18
75%	2	8	10
100%	2	2	4
Total	40	40	80

Ca, Cl, Cu, Mg, Hg, P, Ag, Sn and Zn. Tables IV and V show the statistically significant results for the tooth and amalgam seal, respectively. Non-significant comparisons are not presented due to the large volume of data. The weight percentages of elements in seals of the individual restoration treatments are presented in greater detail in a previous paper (Grossman *et al.*, 1995).

Seal on the tooth cavity surface (Table IV)

Low Ca (17,8-41,2 per cent) and phosphorus (11,2-20,2 per cent) levels were significantly linked to 100 per cent seal whereas a consistent pattern of high Zn (7,3-13,1 per cent) was linked to the 25 and 50 per cent seal. A high Sn content (31,7-59,3 per cent) was linked to 100 per cent sealing which was significantly different from the other seal groups. At three months the 75 per cent seal had a significantly higher Cl content (1,9 per cent) than the other seal classes. This rose to 9,5 per cent in the 100 per cent seal specimens at one year. Combining both times for Mg revealed that the 75 and 100 per cent sealed specimens had a significantly higher Mg content (0,4-0,6 per cent) than the other two sealing classes. Trace amounts of Cu, Hg and Ag were detected on the cut cavity surface. All three elements were present in larger amounts in the better sealing groups.

Seal on the amalgam restoration surface (Table V)

In the three month and combined time group, the Ag content of the 25 per cent and 50 per cent seal was significantly higher (20,9-23,8 per cent) than those of the other two seal groups (3,3-12,4 per cent). Similarly for the same two time

Table IV: *Mean values and (standard deviations) of the % elemental weight composition of marginal seal in the four seal groups as analysed on the tooth. Mean values with the same letter in each time interval are not significantly different at p<0,05,*

Element/time				Percentage seal group				
	25%		50%		75%		100%	
Ca								
3 months	56,3	(7,4)a	56,6	(6,8)a	62,2	(2,6)a	41,2	(12,8)b
1 year	51,1	(11,7)b	49,0	(9,5)b	60,4	(6,0)a	17,8	(16,5)c
Combined time	54,1	(9,8)b	52,4	(9,2)b	60,8	(5,5)a	29,5	(18,7)c
Cl								
3 months	0,9	(0,4)b	1,1	(1,0)b	1,9	(1,5)a	0,7	(0,2)b
1 year	1,7	(2,0)b	1,2	(1,4)b	1,2	(1,1)b	9,5	(5,5)a
Combined time	1,2	(1,4)b	1,2	(1,3)b	1,4	(1,2)b	5,1	(5,9)a
Cu								
Combined time	0,1	(0,4)b	0,1	(0,4)ab	0,4	(0,6)a	0,3	(0,4)ab
Mg								
3 months	0,1	(0,2)b	0,1	(0,2)b	0,4	(0,3)b	1,0	(2,5)a
1 year	0,0	(0,1)b	0,0	(0,1)b	0,3	(0,4)a	0,2	(0,3)ab
Combined time	0,1	(0,2)b	0,1	(0,2)b	0,4	(0,4)a	0,6	(1,8)a
Hg								
3 months	0,3	(0,5)b	0,8	(1,8)ab	0,0	(0,0)b	1,4	(2,1)a
P								
3 months	28,2	(2,8)a	26,8	(3,0)a	27,3	(3,7)a	20,2	(7,5)b
1 year	26,5	(4,9)a	26,9	(3,8)a	27,6	(3,7)a	11,2	(7,4)b
Combined time	27,5	(3,9)a	26,8	(3,5)a	27,5	(3,7)a	15,7	(8,6)b
Ag								
3 months	0,1	(0,4)ab	0,3	(0,8)ab	0,0	(0,0)b	0,5	(0,8)a
Sn								
3 months	4,0	(4,2)b	6,3	(5,6)b	6,1	(5,4)b	31,7	(17,5)a
1 year	9,8	(13,9)b	9,6	(10,2)b	5,3	(5,2)b	59,3	(22,6)a
Combined time	6,4	(9,9)b	8,1	(8,6)b	5,5	(5,2)b	45,5	(24,2)a
Zn								
3 months	10,0	(8,1)a	7,3	(5,5)ab	1,9	(1,7)b	3,1	(1,9)ab
1 year	10,3	(7,9)a	13,1	(8,5)a	4,2	(3,6)b	1,6	(2,1)b
Combined time	10,1	(8,0)a	10,5	(7,8)a	3,8	(3,4)b	2,3	(2,1)b

Please note the comparison is horizontal

intervals, the Hg levels were significantly greater in the 25 per cent and 50 per cent groups (40,6-43,9 per cent) than those specimens in the better sealing categories (6,9-25,9 per cent). The Cu content was significantly higher in the 25 per cent seal material (2,4-2,5 per cent) than the 100 per cent seal group when times were combined and the 75 per cent seal group at three months.

At one year and when times were combined the 100 per cent seal group contained significantly more Cl (3,9-7,9 per cent) than the other groups. A high Sn content (37,2-58,5 per cent) was significantly linked to the 75 per cent and 100 per cent seal groups for all three time groups. Magnesium was significantly greater in the 75 per cent seal group only when times were combined. The 75 per cent seal group had consistently significantly greater amounts of Ca in the seal material (10.4-25,3 per cent) than all other time groups (1,7-4,9 per cent).

DISCUSSION

Previous studies have shown that amalgam, base and cavity varnish affect marginal sealing in the restored cavity. Although the individual contribution of each of these materials to marginal sealing has been previously investigated, the effects of the interaction of these three components on marginal seal formation has not been explored. With time, amalgams are able to form a seal independent of tooth cavity substances (Jodaikin and Grossman, 1984). Previous studies have indicated that the high copper, dispersed phase, admix non-γ_2 Dispersalloy with a Cu_6Sn_6 corrodible phase should leak more than the lathe-cut, conventional low copper γ_2 containing New True Dentalloy (House *et al.*, 1980; Smith, Wilson and Coombe, 1978; Wilson and Smith, 1978; Sarkar, Osborne and Leinfelder, 1982; Fanian, Hadavi and Asgar, 1983; Mahler and Nelson, 1983; Fayyad and Ball, 1984; Craig, 1993; Berry, Nicholson and Troendle, 1994). However, alloys in the same category may vary as to exact formula, particle size composition and manufacturing process (Berry *et al.*, 1994). Additionally, Marek (1992) points out that different parts of the restoration surface may corrode in different ways depending on the prevailing interfacial conditions. Both the above could contribute to the previously conflicting results on alloy type, corrosion and marginal leakage.

Well documented reports implicate bases under an amalgam alloy with increased marginal leakage *in vitro* (Swartz and Phillips, 1962; Norman, Swartz and Phillips, 1963; Wilson and Smith, 1978; Manders, Garcia-Godoy and Barnwell, 1990; Rabchinsky and Donly, 1992). Marginal leakage has been linked in some cases with the disintegration of calcium hydroxide cements *in vivo* (Going, 1972; Gourley and Rose, 1972; Pereira *et al.*, 1990).

The ability of a varnish to effectively seal an amalgam restored cavity appears to depend on the type of alloy and varnish, its solubility, the time elapsed between varnish placement and leakage assessment and the number of layers of varnish applied (Eames and Hollenback, 1966; Smith *et al.*, 1978; Murray, Yates and Williams, 1983; Newman, 1984; Sneed, Hembree and Welsh, 1984; Gottlieb, Retief and Bradley, 1985; Ben-Amar *et al.*, 1986; Newman and Szojka, 1986; Powell and Daines, 1987; Kelsey and Panneton, 1988; Liberman *et al.*, 1989; Fitchie *et al.*, 1990). Two coats of a low solubility varnish placed against a conventional amalgam and tested immediately gives the best seal. However contradictory sets of experimental results complicate straightforward conclusions on this matter (Mazer, Rehfeld and Leinfelder, 1988).

Table V: *Mean values and (standard deviations) of the % elemental weight composition of marginal seal in the four seal groupings as analysed on the amalgam. Mean values with the same letter in each time interval are not significantly different at p,0,05.*

Element/time	**Percentage seal group**							
	25%		**50%**		**75%**		**100%**	
Ca								
3 months	2,3	(5,3)b	4,1	(4,1)b	25,3	(4,5)a	4,9	(1,9)b
1 year	1,7	(2,7)b	3,7	(7,1)b	10,4	(8,5)a	1,8	(1,7)b
Combined time	2,0	(4,4)b	3,9	(5,9)b	13,4	(9,9)a	3,4	(2,4)b
Cl								
3 months	0,4	(0,7)b	0,4	(0,6)b	3,1	(4,1)a	0,0	(0,0)b
1 year	1,6	(3,4)b	1,6	(3,3)b	1,1	(3,2)b	7,9	(4,6)a
Combined time	0,9	(2,3)b	1,1	(2,5)b	1,5	(3,4)b	3,9	(5,2)a
Cu								
3 months	2,5	(1,3)a	1,8	(0,9)ab	0,4	(0,3)b	0,9	(0,3)ab
Combined time	2,4	(1,3)a	1,7	(0,9)ab	1,9	(1,4)ab	0,9	(0,3)b
Mg								
Combined time	0,0	(0,1)b	0,1	(0,2)b	0,3	(0,5)a	0,1	(0,1)b
Hg								
3 months	43,9	(7,1)a	41,0	(5,8)a	6,9	(7,4)c	25,9	(3,7)b
Combined time	41,3	(9,9)a	40,6	(9,1)a	24,2	(18,8)b	25,9	(3,4)b
Ag								
3 months	23,8	(4,8)a	20,9	(3,2)a	3,3	(3,9)c	11,7	(1,1)b
Combined time	22,2	(6,2)a	20,8	(5,1)a	12,4	(10,1)b	12,1	(1,3)b
Sn								
3 months	22,2	(8,5)b	25,5	(8,2)b	58,5	(8,6)a	52,8	(4,1)a
1 year	30,7	(17,0)ab	25,1	(13,1)b	37,2	(25,0)ab	48,4	(4,6)a
Combined time	25,7	(13,4)b	25,3	(11,1)b	41,5	(24,1)a	50,6	(4,7)a

Please note the comparison is horizontal

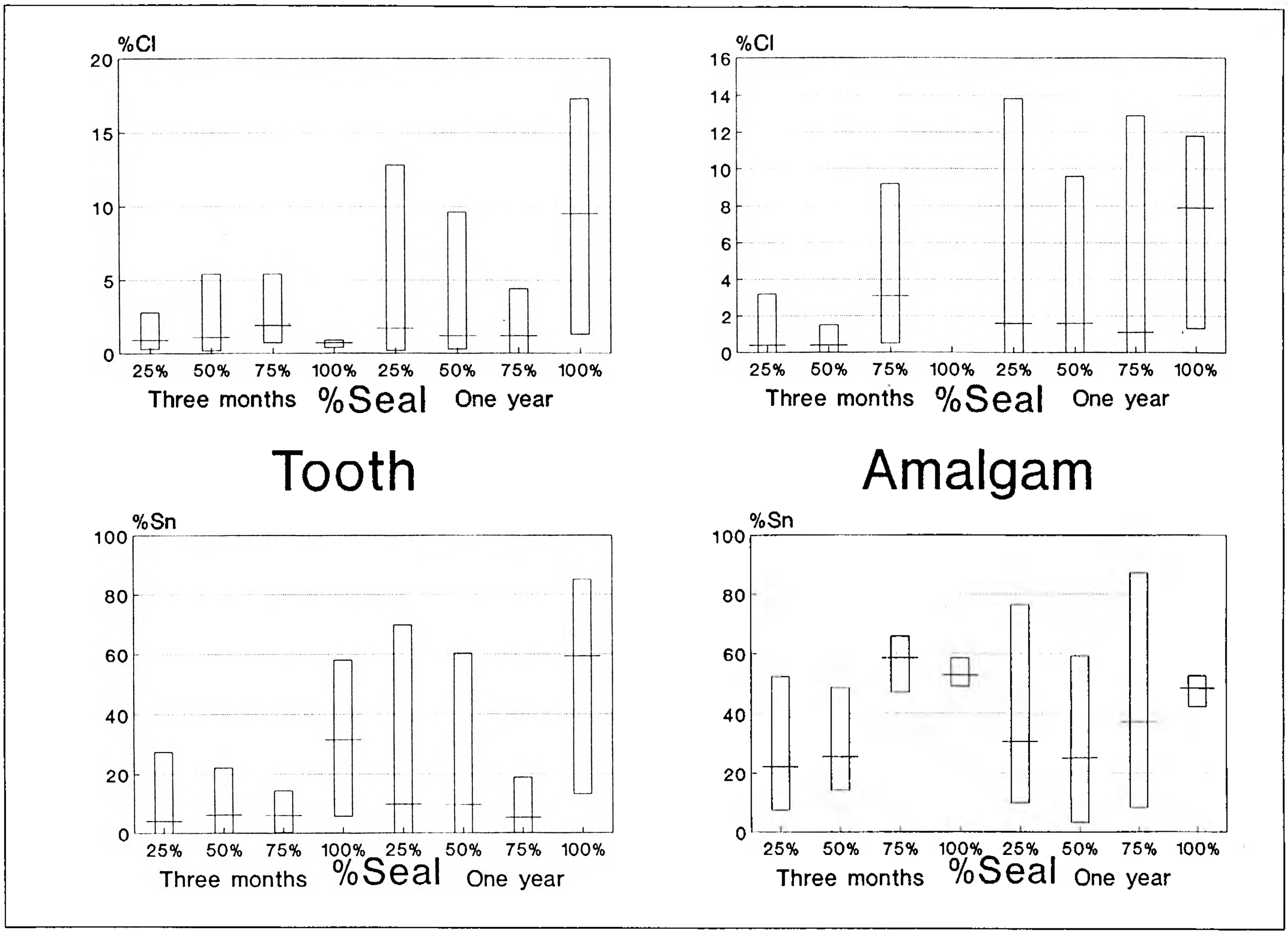

Fig. 2: **Maximum and minimum values for Cl and Sn obtained in the seal material as analysed on the tooth and amalgam in the four seal groups. The bar indicates the mean value.**

The marginal leakage results of this study indicate that the link between amalgam type and leakage has been followed in that New True Dentalloy specimens seal better overall than Dispersalloy. However, when the latter is combined with a calcium hydroxide base, the poor sealing ability of high copper amalgam is compensated for and, as a result, the calcium hydroxide paste based Dispersalloy specimens sealed consistently well at all time intervals. As shown in previous studies the bases in this investigation enhanced marginal leakage, except for calcium hydroxide paste base specimens where sealing was promoted. Unvarnished cavities sealed better at three months except in combination with Dycal.

The elemental analysis study partially confirms the long held belief that Sn/Cl complexes play an important role in the sealing mechanism at the amalgam-tooth interface (Jodaikin and Goldstein, 1988). Both these elements were present in significantly greater amounts in the tooth and amalgam marginal seals of the better sealing specimens. However, these two elements show a large standard deviation and a wide range of minimum and maximum values (Fig. 2). Thus some treatment groups containing large amounts of Sn and Cl in the seal showed poor sealing at one year (Grossman and Matejka, 1993), such as unvarnished Dycal+Cu(low), Kalz+Cu(low) and varnish+ZnO+Cu(low). Conversely a good seal is present in the one year unvarnished Calc+Cu(high), varnish+Cu(high) and varnish+Calc+Cu(low) specimens where lower Sn and Cl values occur (Grossman *et al.*, 1995).

The significant linking of Mg to the better sealed specimens is an unexpected and interesting phenomenon. Magnesium is readily dissolved from the calcified tissue by body fluids (Jenkins, 1978). In 1971 Inoue (Hals, Tveit and Tøtdal, 1988) demonstrated increased Mg in the dentine of abraded teeth. Whether cavity cutting followed by incubation in 1 per cent NaCl could result in a similar movement of Mg into the marginal seal material or whether active uptake of Mg is occurring can only be speculated upon.

The reduction in concentration of Ag and Hg in the amalgam specimens is an indication of the increasing deposition of seal material on these two surfaces. This topic has been dealt with previously (Witcomb, Grossman and Jodaikin, 1987). The reduction of Ca and P in the tooth seal analyses is probably due to the same reason although demineralization of tooth structure adjacent to the restoration is a possibility. However, a review of X-ray micro-analysis of dentine (Hals *et al.*, 1988) point out conflicting results in Ca and P values in dentine wall lesions where microradiographs showed increased radio-opacity. Reduced Ca and P values and increased amounts of Sn and Zn had been exhibited in such areas adjacent to amalgam restorations. Conversely areas adjacent to silicate restorations displayed increased Ca and P values together with varying concentrations of elements such as Zn derived from the restorative material. The high levels of Ca present in the 75 per cent sealing amalgam specimens in our study can be attributed mainly to the unvarnished+calc+Cu(high) specimens (Grossman *et al.*, 1995).

McTigue *et al.*, (1984) have noted that the oxidation process of the CuSn phase leads to the formation of relatively soluble Cu complexes. Leaching of these complexes is the reason for the absence of Cu-containing corrosion products in their study on Dispersalloy corrosion. Similar leaching could be responsible for the reduced levels of Cu in the seal material as analysed in both configurations in our study. Additionally, reduced levels of Cu in seal material analysed on the amalgam could be an indication of increasing amounts of seal material being deposited on the restoration surface.

Both amalgams used in this study contain small amounts of Zn (Grossman *et al.*, 1995). The Zn-containing bases have also contributed to the pool of Zn at the tooth-amalgam interface. Previous studies related to our investigation have noted the mobility and/or the high initial dissolution rate of Zn from the amalgam (Sarkar *et al.*, 1981; Brune, 1981; Palaghias, 1986; Marek, 1988; Patsurakos and Moberg, 1990). Hals *et al.*, (1988) refers to the presence of Zn commonly found in the tooth adjacent to Zn-containing restorative materials. The low Zn ratios associated with good sealing on tooth specimens could be due to either Zn inhibiting sealing, or an indication of increasing amounts of seal material being deposited over tooth material invaded by Zn. Although Zn proved to be significantly linked to leakage when the seal material was analysed on the tooth, the Zn content of seal material on the amalgam seemed to have no influence on sealing.

A problem arising from the grouping of specimens into classes based on sealing has meant that the results could be interpreted as being heavily biased towards those elements primarily associated with the calcium hydroxide based restoration treatments as these specimens are the only ones which sealed completely over the one year experimental period. To investigate this, smaller subgroups were similarly analysed using selection criteria such as varnish, amalgam or base type. The seal elements were then compared to the amount of seal achieved as previously. The same basic trends (results not shown) were apparent as reported above. Dycal based specimens remained paradoxical throughout with poor sealing in the long term (Grossman and Matejka, 1993) while the elemental composition of the marginal seal indicated a well formed seal (Grossman *et al.*, 1995).

It is suggested that a period of one year is not long enough to fully explore the cascade of events which could lead to sealing at the amalgam-tooth interface following restoration. Five years might be more realistic in view of the fact that barely half of the specimen combinations showed 25 per cent or less seal after one year. The practicalities of such an undertaking in terms of obtaining suitable teeth would be daunting. A major concern in restorative dentistry is the inability of dental materials to adapt to and seal, margins of cavities. Our results show those chemical elements essential for speedy amalgam cavity sealing. Such information has vast clinical implications in terms of restoration longevity through reduced marginal leakage time.

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