Penetration of ions from silicate cement restorations into Copalite® – covered cavity walls

ANNE BJØRG TVEIT & EINAR HALS

Department of Cariology, School of Dentistry, University of Bergen, Norway

Tveit, A.B. & Hals, E. Penetration of ions from silicate cement restorations into Copalite©-covered cavity walls. *Acta Odont. Scand.* 36, 15–24

This study aimed to assess the effect of silicate cement on Copalite®-covered cavity walls in extracted human teeth. Class V cavities were prepared in 24 premolars and filled with silicate cement (Bio-Trey®). Four cavities were unlined, the rest of the cavities were lined with 1 or 2 layers of Copalite before insertion of the restorations. After 6 months, 70–100 μm thick longitudinal sections of the teeth were studied by polarized light microscopy, microradiography and electron probe microanalysis. When imbibed in water or quinoline, a subsurface zone of altered birefringence was noticed in nearly all cavity walls. Nearly half of the cavity walls in the experimental groups showed a surface zone of increased radiopacity. In a few instances a subsurface radiolucent zone was present. By electron probe microanalysis F (0,4–3% by weight), Zn (1–4%) and Al (0,2–6%) were measured in the outer 10–60 μm of the cavity walls. The study shows that even with a double layer of Copalite, known to prevent microleakage, a desirable uptake of F and Al from silicate restorations into cavity walls can take place. Copalite does not prevent a phosphoric acid effect on the cavity walls.

Key-words: Filling material; varnish; fluoride

Anne Bjørg Tveit, School of Dentistry, Årstadvei 17, N–5000 Bergen, Norway

The main functions of cavity varnishes are to prevent the penetration of deleterious constituents of restorative materials into the dentin and the pulp and to reduce microleakage between the restoration and the cavity walls. Cavity varnishes consist mainly of natural and synthetic resins dissolved in an organic solvent such as chloroform, acetone or ether. A cavity varnish applied to the cavity walls forms a film which acts as a semipermeable membrane (12).

The varnish, Copalite®, does not entirely prevent the passage of phosphoric acid liberated from silicate cement into the enamel and dentin, but greatly reduces it (18, 19). The latter study was undertaken *in vivo*, the former *in vitro*. Copalite tends to inhibit the penetration of fluoride into enamel and dentin (13, 16, 17). The effect of silicate cement on Copalite-covered enamel was studied by Norman et al. (11) in experiments of 2 weeks' duration. The uptake by enamel of fluoride

Received for publication, September 12, 1977

released from the silicate cement was reduced by approximately 60 per cent compared with cases where Copalite had not been used. On these grounds it has been recommended that when a varnish is used in connection with silicate cement, it should be removed from the enamel walls (12). Forsten and Paunio (4) studied the uptake of fluoride released from Copalite-coated silicate cement into synthetic hydroxy-apatite. During the first 3 weeks the uptake was greater from the uncoated than from the coated silicate cement, but from the 4th week on there was no difference between them. These results indicate that the varnish has the effect of reducing the initial release of fluoride from silicate cement, while the long-term effect is about the same with or without the varnish.

The permeability of Copalite has also been assessed in experiments in which radioactive isotopes and dyes have been used (1). The varnish was found to be effective in preventing marginal and dentin penetration in unfilled cavities and in cavities filled with silicate cement. Using water vapour, Eriksen and Nordbø (3) found that Copalite had lower permeability values than the liner Tubulitec®. Three coats of varnish provide a more protective barrier against phosphoric acid than a single coat (19). Naturally, the efficiency of the film of varnish in preventing acid penetration will depend upon its continuity.

The aim of the present study was to assess the topographical distribution in Copalite-covered cavity walls of F, Al and Zn ions released from silicate cement restorations. The topographical distribution of Ca, P, Mg and Na ions will also be discussed.

MATERIAL AND METHODS

A total of 24 extracted human premolars were used in this investigation. The teeth were stored in 10 per cent neutral formalin and were not allowed to dry during any stage of the experiment.

Class V cavities were prepared on clinically sound buccal surfaces using an air rotor with adequate water cooling. The cavity walls were finished with diamond (80.000 rpm) and finishing burs (15.000 rpm).

After rinsing with water the cavities were dried and silicate cement (Bio-Trey®) was inserted. In 8 cavities a single coat of Copalite was applied with a cottonwool pellet before insertion (Group I), while in 8 cavities two coats were used (Group II). Four cavities served as control of the effect of the constituents of silicate cement on cavity walls not coated with Copalite (Control group I). The cellophan strips used as matrices for the restorations were removed after 5 minutes and the buccal surfaces covered with petroleum jelly. Excess filling material was removed after 15 minutes with petroleum jelly-covered sandpaper discs at low speed. Following removal of the petroleum jelly the teeth were stored for 6 months (2 teeth for 3 months only) at 37 °C in a physiological saline solution which was changed weekly. Longitudinal sections, 70–100 μm thick, passing through the cavities, were then prepared. To find out whether smearing of the filling material onto the surfaces of the sections occurred, the restorations were carefully removed from 2 cavities coated with 1 layer and 2 cavities coated with 2 layers of Copalite before sectioning of the teeth (Control group II).

The sections from the experimental groups and Control group I were examined by polarized light microscopy, microradiography and electron probe microanalysis, while the sections from Control group II were examined by electron probe microanalysis only.

The sections to be used with the latter method were covered with a thin layer of carbon and examined in an ARL electron microprobe operated at 10 and 20 kV and 0,2–0,67μA. In several areas of each specimen analyses were carried out for Ca, P, Mg, Al, Zn, F and Na using the $K\alpha$ emission of each element. Pulse height discrimination was used for the analysis of F. Linear scans were made at right angles to the cavity walls. X-ray images showing the distribution of some of the elements were displayed on an oscilloscope and photographed with a Polaroid® camera. Weight percentages of the element in each specimen were calculated by comparison with the X-ray emission of the following minerals which previously have been found appropriate for the elements in question (7, 9):

Standard	*Element*
Apatite	Ca P F
Biotite	Mg Na Al
Sphalerite	Zn

The electron probe techniques used in this study have been more fully described by Halse (8).

RESULTS

No significant differences were observed between groups I and II, regardless of the method used. These groups are, therefore, described as a whole.

Polarized light microscopy

Sections imbibed in destilled water revealed no changes in the dentin neither in the experimental groups nor in Control group I. In the enamel, however, a reduced negative or pseudo-isotropic birefringent subsurface zone was observed in 20 out of 25 cavity walls in the experimental groups (Fig. 1) and in 6 out of 8 cavity walls in Control group I. When the sections were imbibed in quinoline the same area of the enamel in nearly all instances showed a positive or pseudo-isotropic zone (Fig. 2).

Microradiography

Experimental groups: Enamel changes occured in 12 out of 26 cavity walls. (Some sections were spoiled during the electron probe analysis.) Usually, a surface zone of

Figs. 1–9 refer to cavities lined with 1–2 layers of Copalite, filled with silicate cement and stored in weekly changed saline at 37 °C for 6 months.

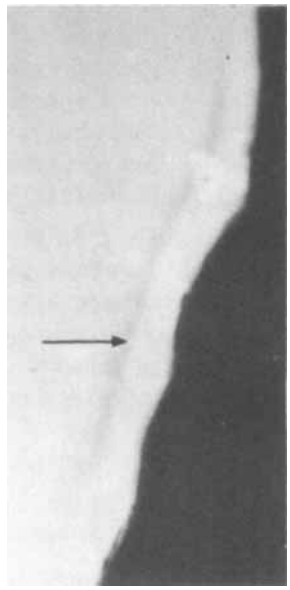


Fig. 1. Enamel cavity wall. One layer of Copalite. Narrow subsurface zone showing pseudo-isotropy (arrow). Polarized light. Section imbibed in destilled water. Original magnification 100 x.

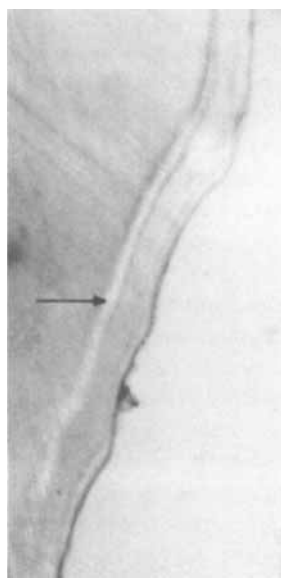


Fig. 2. Same section as in Fig. 1, imbibed in quinoline. Polarized light. The subsurface zone appears as a positively birefringent zone bordered by pseudo-isotropic lines (arrow). Original magnification 100 x.

greater radiopacity than normal enamel was noticed. In 3 instances a slightly radiolucent, narrow subsurface zone was observed. These changes were not so distinct in the experimental groups as in the control groups. In 12 out of 26 cavity walls a 10–20 μm wide surface zone of the dentin showed increased radiopacity compared to normal dentin (Fig. 4) and in 2 instances a subsurface radiolucent zone was also observed.

Control group I: In the enamel a narrow, subsurface radiolucent zone was visible in 2 out of 8 cavity walls. All cavity walls showed a 10–20 μm wide dentinal surface zone of increased radiopacity. In 4 instances a subsurface radiolucent zone was present (Fig. 3).

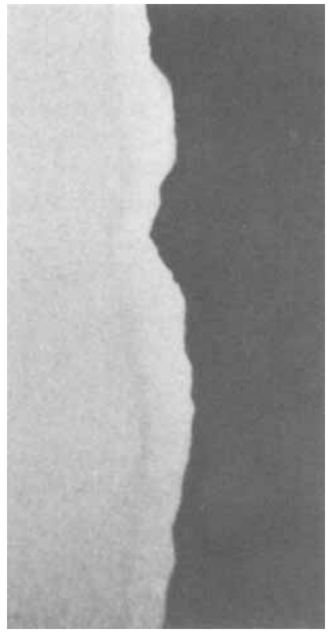


Fig. 3. Surface zone of greater radiopacity than normal enamel. Subsurface radiolucent zone. Microradiograph. Original magnification 100 x.

Electron probe microanalysis

Experimental groups: The concentration profiles of Ca, P, Mg and Na in the experimental groups most frequently appeared as largely horizontal lines. In approximately one third of the specimens the Ca, P and Mg scans showed a surface or subsurface depression of varying width mainly indicating a 10% lowering of the concentration values (Figs. 5, 7). A few Mg-profiles showed a slight elevation near the cavity wall. In the dentin most of the Na profiles showed an increased concentration near the cavity wall (Fig. 6). Further data concerning Ca, P, Mg and Na are given in Table I. A survey of data on the concentrations of Zn, Al and F in the dentin enamel walls is given in Table 2. The concentrations were generally slightly higher in Group I than in Group II, but the differences were not significant, and the groups are, therefore, described as a whole. Increased concentrations of Zn – slightly higher in the dentin than in the enamel – were observed in nearly half of the scans. The concentration peaks of these zones were 10–20 μm wide (Fig. 7). In almost all instances a marked elevation of the F and Al profiles near to the cavity wall was noticed (Figs. 5, 6). This zone was considerably wider in the F scans than in the Al scans, and the concentration of the elements was 2–3 times higher in the dentin than in the enamel. Figs. 8 and 9 show elemental X-ray images of the distribution in the dentin wall of Al and Zn, respectively.

Control group I: The configuration of the Ca, P and Mg profiles and the concentrations of Zn, Al and F could not be seen to differ to any significant degree from those of the experimental material.

Control group II: It was not possible to observe any differences between the concentration profiles from this group and the experimental groups.

DISCUSSION

It has been shown that the results of the electron probe microanalysis have not been influenced by any smearing of particles from the silicate material onto the surface of the

Table 1. *Concentration profiles of Ca, P, Mg and Na in enamel/dentin of cavity walls. Electron probe microanalysis*

Ratio $\frac{\text{observed number of profile variants*}}{\text{total number of relevant scans**}}$

Experimental groups

Elements	Profile as horizontal line	Depression of profile near cavity wall	Elevation of profile near cavity wall
Ca } P }	29 : 41	12 : 41	
Mg	28 : 48	17 : 48	3 : 48
Na	13 : 20 (enamel only)		14 : 20 (dentin only)
Control group I			
Ca } P }	14 : 23	9 : 23	
Mg***	1 : 4	3 : 4	
Na	2 : 4		2 : 4

* Three variants, see upper part of table.
** i.e. scans of Ca, P, Mg and Na, respectively.
*** Observation from ground surfaces.

sections during processing of the sections. Because most of the fillings were lost during preparation of the sections the possibility of fluorescence at the cavity walls could be excluded. Such fluorescence, if present, would have invalidated the quantitative estimation of the elements in this area.

Previous short term studies (16, 18, 19) have shown that Copalite will greatly reduce the penetration of phosphoric acid from silicate into enamel and dentin. In our (long-term) studies, however, no noteworthy difference between the effect of the acid on coated or uncoated cavity walls was apparent. The concentration profiles of the Ca, P and Mg indicate a demineralization in one third of the scans in the experimental and control groups. There was always a correspondance between the narrow radiolucent zone in the enamel wall observed in the microradiographs, and the subsurface reduced negative or pseudo-isotropic zone observed when the specimens were imbibed in water. These observations and those made in specimens imbibed in quinoline indicate demineralized areas. The subsurface zone (Fig. 2) corresponds to the «positive zone in quinoline» in natural and artificial caries, and is explained by the presence of narrow spaces, unpenetrable by the large quinoline molecules but penetrable by water molecules (14, 15). However, unlike the situation in natural and artificial caries, the corresponding zone in the present material in some instances showed pseudo-isotropy or reduced negative birefringence also when examined in water. Probably the zone contains a larger number of the described small spaces than the corresponding zone in natural and artificial caries. In sections immersed in water a positive form birefringence may, therefore, compensate or reduce the effect of the negative birefringence of the enamel apatite, resulting in pseudo-isotropy or reduced negative birefringence, respectively. In quinoline the sections must reveal

Table 2. *Concentrations of Zn, Al and F in Copalite-covered or uncovered cavity walls adjacent to silicate cement restorations. Electron probe microanalysis*

Element	Experimental groups					Control group		
	Concentration		Depth of increased concentration	Frequency of scans in which observed		Concentration	Depth of increased concentration	Frequency of scans in which observed
	Range	Average				Average		
Zn	Enamel: 1%–4%	2%	10–20 μm (concentration peak)	26:51	10:25			
	Dentin: 2%–3.5%	2.8%	10–20 μm (concentration peak)		16:26	Dentin 2.5%	10–20 μm	3:3
Al	Enamel: 0.2–2.5%	0.9%	10–20 μm	43:46	21:24	Enamel: 1.5%	10–20 μm	4:4
	Dentin: 0.3–6%	2.4%	10–20 μm		22:22	Dentin: 2.5%	10–20 μm	7:7
F	Enamel: 0.4%–2%	0.8%	15–60 μm (average 30 μm)	45:49	23:24	Enamel: 1%	Average 40 μm	6:6
	Dentin: 0.8%–3%	1.8%	15–100 μm (average 50 μm)		22:25	Dentin: 2.1%	Average 80 μm (observed from 7 scans)	14:14

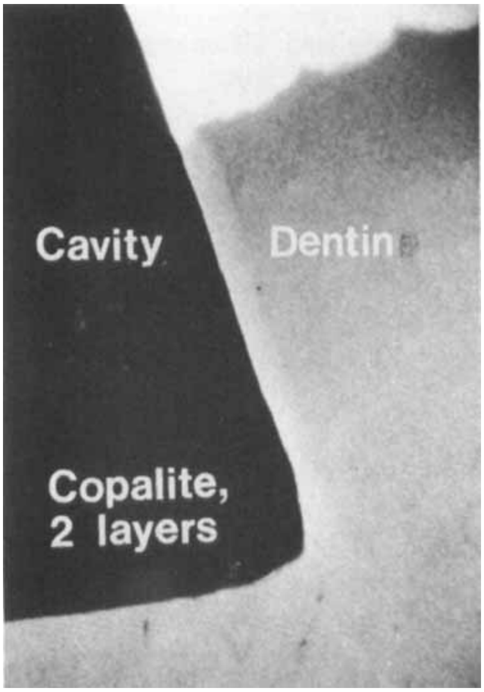


Fig. 4. Dentin cavity wall. Two layers of Copalite. Increased radiopacity relative to normal dentin in the surface zone. Microradiograph. Original magnification 100 x.

psuedo-isotropy or positive birefringence depending on the number of spaces.

By deduction from figures given by Darling (2) the zone examplified by Fig. 1 may contain > 5% spaces. This is slightly higher than in the corresponding zone described by Hals (5) in a study of the effect of silicate cement on uncoated enamel walls. However, this slight discrepancy may be explained by a longer experimental time in the present study, or a thinner silicate mix. Apparently, phosphoric acid from the silicate cement has penetrated the Copalite film in more than half of the specimens.

The zones recorded on the microradiographs could not always be explained by the electron probe analysis. The reason for this has been discussed previously (6).

In our experiments no significant reduction of fluoride uptake was observed. This is not in accordance with the data reported by Norman et al. (11), but conforms with the result of Forsten and Paunio (4), who, however, showed that Copalite had a definite reducing effect on the initial release of F.

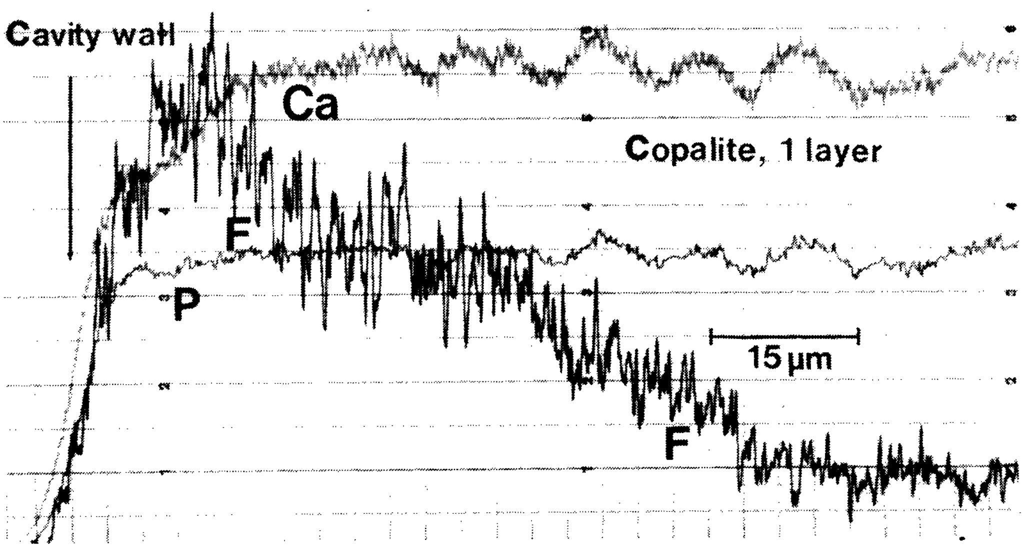


Fig. 5. Electron probe. Linear scans through dentin cavity wall. One layer of Copalite. Surface depression of Ca and P profiles, indicating ~ 10 % reduction of concentration values. High concentration of F (~ 3 %) at surface of dentine wall, decreasing to normal values at a distance of 65 µm from the surface.

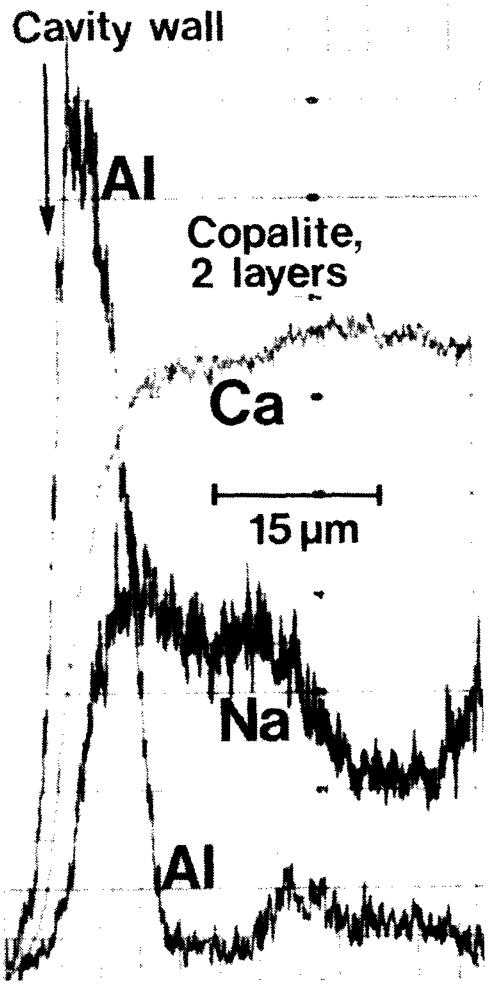


Fig. 6. Electron probe. Linear scans through dentin cavity wall. Two layers of Copalite. Surface lowering of Ca profile, surface elevation of Na and Al profiles.

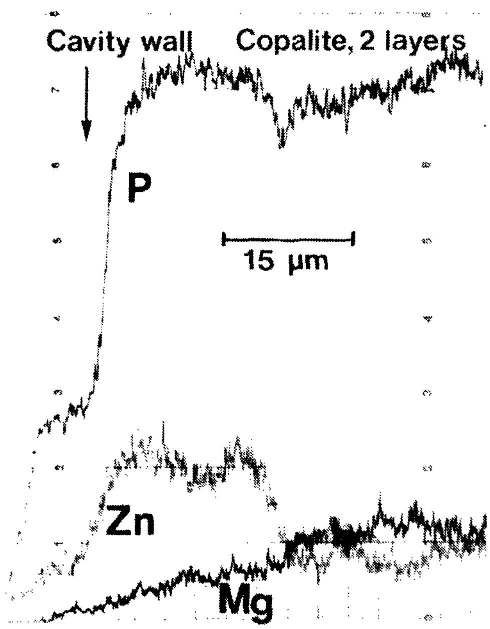
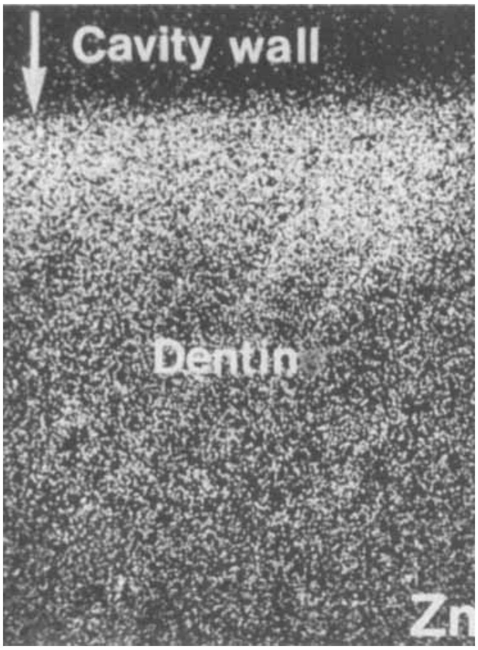


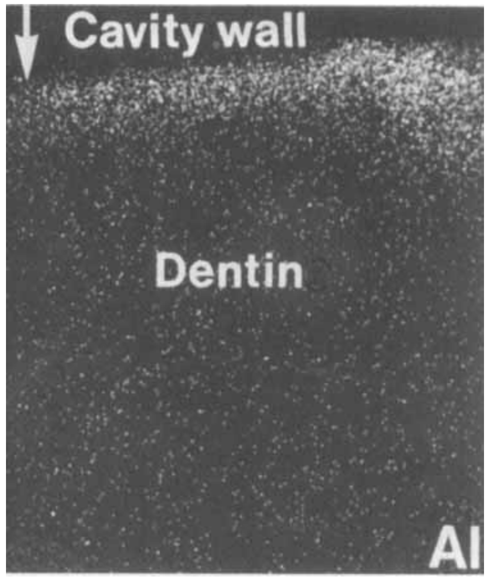
Fig. 7. Electron probe. Two layers of Copalite. Linear scans through dentin cavity wall. Subsurface depression of P, surface depression of Mg, and surface elevation of Zn profile.

Copalite does not seem to prevent uptake of Zn and Al to any registrable degree. In all probability the Zn ions have contributed to the zone of increased radiopacity in the cavity walls. Since the mass absorption coefficient increases as the fourth power of the atomic number, the presence of more than 0,1% Zn in the surface layer will significantly increase its opacity to soft x-rays (7). However, the width of such zones could not in all cases be explained by electron probe analysis. Halse and Hals (9) have shown that the concentration of Zn decreased gradually at a varying distance up to 400 µm into the dentin. Probably our registrations (150–200 µm into the dentin) refer to a concentration peak near the cavity wall, while a slight uptake may go deeper into the tissue.

The Mg concentration profiles showed a varying configuration. This supports previous findings (20, 9). No explanation is offered for the presence of the Na ions in the specimens. It should, however, be mentioned that the concentration of Na in Bio-Trey powder is 5,8% (10). It is generally assumed that the low frequency of secondary caries in silicate-filled teeth is due to uptake of fluoride in cavity walls. Through its tendency to form complexes with F, Al may possibly enhance the cariostatic effect of F (9). The importance of the uptake of Zn is uncertain.

The study indicates that before insertion of silicate restorations the cavity walls should be coated with varnish in order to reduce microleakage. Since Copalite varnish does not





Figs. 8–9. Elemental X-ray images showing the distribution of Zn and Al in dentin cavity wall.

significantly hinder the uptake of F and Al by the cavity walls, it should not be removed from the enamel walls. However, when applied as a double layer below silicate restorations, Copalite does not always prevent a phosphoric acid effect on the cavity walls.

Acknowledgement. The electron probe analyses were carried out at the Institute of X-ray Crystallography, Technical University of Norway, Trondheim.

REFERENCES

1. Barber, D.B., Hines, I. & Massler, M. Penetration of isotopes through liners and bases under silicate cement restorations. J. Am. Dent. Assoc. 1962, 65, 786–796
2. Darling, A.I. Studies of the early lesion of enamel caries with transmitted light, polarized light and microradiography. Its nature, mode of spread, points of entry and its relation to enamel structure. Brit. Dent. J. 1958, 105, 119–135
3. Eriksen, H.M. & Nordbø, H. Permeability of different lining materials. Scand. J. Dent. Res. 1974, 82, 442–447
4. Forsten, L. & Paunio, I.K. Fluoride release from varnish-coated silicates and from cavity liners and fissure sealants. Scand. J. Dent. Res. 1973, 81, 513–517
5. Hals, E. The structure of experimental in vitro lesions around silicate fillings in human teeth. Archs. Oral. Biol. 1975, 20, 283–289
6. Hals, E. & Halse, A. Electron probe microanalysis of secondary carious lesions associated with silver amalgam fillings. Acta Odont. Scand. 1975, 33, 149–160
7. Hals, E. & Selvig, A. Correlated electron probe microanalysis and microradiography of carious and normal dental cementum. Caries. Res. 1977, 11, 62–75
8. Halse, A. An electron microprobe investigation of the distribution of iron in rat incisor enamel. Scand. J. Dent. Res. 1972, 80, 26–39
9. Halse, A. & Hals, E. Electron probe microanalysis of secondary carious lesions adjacent to silicate fillings. Calcif. Tiss. Res. 1976, 21, 183–193
10. Helgeland, K. & Leirskar, J. Silicate cement in a cell culture system. Scand. J. Dent. Res. 1973, 82, 251–259
11. Norman, R.D., Platt, J.R., Phillips, R.W. & Swartz, M.L. Additional studies on fluoride uptake by enamel from certain dental materials. J. Dent. Res. 1961, 40, 529–537
12. Phillips, R.W., Swartz, M.L. & Norman, R.D. Materials for the practicing dentists. The C.V. Mosby Company, Saint Louis, 1969, pp15 and 21
13. Platt, J.R. & Norman, R.D. Fluoride uptake by

intact enamel from certain dental materials. J. Dent. Res. 1960, 752 (Abstract)
14. Poole, D.F.G., Darling, A.J., Mortimer, K.V. & Ollis, W.D. Molecular sieve behaviour of normal and carious enamel. Archs. Oral Biol. 1961, 5, 251–273
15. Silverstone, L. M. Observations on the dark zone in early enamel caries and in artificial caries-like lesions. Caries Res. 1967, 1, 261–274
16. Swartz, M.L. & Phillips, R.W. Permeability of cavity liners to certain agents. J. Dent. Res. 1960, 39, 1232–1239
17. Swartz, M.L., Phillips, R.W. & Chamberlain, N. Continued studies on the permeability of cavity liners. J. Dent. Res. 1962, 41, 66–74
18. Swartz, M.L., Phillips, R.W., Norman, R.D. & Niblack, B.F. Role of cavity varnishes and bases in the penetration of cement constituents through tooth structure. J. Prosthet. Dent. 1966, 16, 963–972
19. Swartz, M.L., Niblack, B.F., Alter, E.A., Norman, R.D. & Phillips, R.W. In vivo studies on the penetration of dentin by constituents of silicate cement. J. Am. Dent. Assoc. 1968, 76, 573–578
20. Takuma, S., Ogiwara, H. & Suzuki, H. Electron-probe and electron microscope studies of carious dentinal lesions with a remineralized surface layer. Caries. Res. 1975, 9, 278–285