Elements in Marginal Seals at Amalgam-Tooth Interfaces
E.S. Grossman, M.J. Witcomb and A. Jodaikin
J DENT RES 1986 65: 998
DOI: 10.1177/00220345860650072001

The online version of this article can be found at:
http://jdr.sagepub.com/content/65/7/998

Published by:
SAGE
http://www.sagepublications.com

On behalf of:
International and American Associations for Dental Research

Additional services and information for Journal of Dental Research can be found at:
Email Alerts: http://jdr.sagepub.com/cgi/alerts
Subscriptions: http://jdr.sagepub.com/subscriptions
Reprints: http://www.sagepub.com/journalsReprints.nav
Permissions: http://www.sagepub.com/journalsPermissions.nav
Citations: http://jdr.sagepub.com/content/65/7/998.refs.html

>> Version of Record - Jul 1, 1986
What is This?
Elements in Marginal Seals at Amalgam-Tooth Interfaces

E.S. GROSSMAN¹, M.J. WITCOMB², and A. JODAIKIN³

¹MRC/University of the Witwatersrand Dental Research Institute and ²Electron Microscope Unit, University of the Witwatersrand, Johannesburg, South Africa; ³Isotope Department, The Weizmann Institute of Science, Rehovot, Israel

X-ray micro-analysis was used to determine the elemental composition of the tooth, amalgam, and base material of ten aged, amalgam-restored, posterior teeth after they had been fractured across the amalgam-tooth interface. Subsequently, replicas were used to remove marginal seal material from each interface for examination by x-ray micro-analysis. The results show that P and Sn were present in all marginal seal material analyzed. Ca, Cl, Cu, Fe, S, and Zn were present in the marginal seals of various numbers of specimens.


Introduction.

A crevice exists at the freshly condensed amalgam-tooth interface, or it may develop after varnishes under aged dental amalgam restorations craze or are lost (Jodaikin, 1981). This crevice is sufficiently wide for micro-organisms and oral fluids to pass along the cavity walls as marginal leakage which facilitates the development of dental caries at the interface and thereby accounts for the majority of amalgam restoration replacements (Richardson and Boyd, 1973).

Marginal leakage does not proceed indefinitely since, with time, the crevice becomes sealed. The formation of this seal is not a simple process. The various materials used to prepare the cavity and restore the teeth, along with their interactions, oral physiological factors, and other causes, such as diet, may determine the type of seal and the speed with which it develops. The identification and characterization of distinctive features of the marginal seal, in order that the roles of the contributory factors may be better understood, may aid clinical studies directed toward determining the effects of various types of marginal seal on the longevity of amalgam restorations (Jodaikin and Grossman, 1984).

One method of describing the marginal seal is through the identification of elements within the seal by means of x-ray micro-analysis (Wei and Ingram, 1969; Holland and Asgar, 1974; Sarkar et al., 1981). However, studies so far have been on bulk specimens where electron-beam scattering on and in the sample can cause generation of spurious x-rays from material bordering the seal. To overcome this interference effect, the present study used a low-atomic-weight material to replicate the specimen surface as well as to extract marginal seal material from the interface for analysis.

Materials and methods.

Ten posterior teeth with occlusal amalgam restorations, which were without secondary caries and which had been extracted for periodontal reasons, were rinsed briefly under running tap water and dried at 37°C. The history of specimens was not known except that the restorations had been in mouths for more than six months. The types of amalgams used were also not known, but our study does allow for characterization based on Cu values. The roots of the teeth were mounted in resin blocks and the crowns partially sectioned on three sides with a low-speed, water-cooled, diamond blade, rotary saw (Isomet, Buehler, Ltd., Evanston, IL 60044) in order to facilitate a fracture perpendicular to the amalgam-tooth interface. The fractured surface was lightly coated with high-purity carbon and examined at 20 kV in a Cambridge S4 scanning electron microscope (Cambridge Instruments Ltd., Rustat Road, Cambridge CB1 3QH, U.K.), equipped with an EDAX 707B energy-dispersive x-ray analyzer (EDAX Laboratories, P.O. Box 135, Prairie View, IL 60069). X-ray analysis was carried out on the tooth, base, and amalgam. Analysis conditions were: a live counting time of 200 seconds and count rate adjusted to 1000 counts per second.

Subsequently, a dam of silicone material (GC Exaflex®, regular type, G-C Dental Industrial Corp., Japan) was built in the fractured area to surround completely, but not encroach on, the amalgam-tooth interface. A 25% solution of nitrocellulose in amyl-acetate (Ernest F. Fullam, Inc., Schenectady, NY 12302) was dripped carefully into the dam and allowed to dry at a temperature of 18°C. The solid nitrocellulose replica was then stripped from the tooth, secured to a graphite mount, and coated with high-purity carbon (Fig. 1). The extracted seal material found along the replicated tooth-amalgam interface was then analyzed (Fig. 2). In order to obtain reference energy spectra, we also undertook control analyses of the nitrocellulose replicating material, Copalite® varnish (Copalite®, Cooley & Cooley, Houston, TX), zinc oxide eugenol (Kalzinol®, A.D. International Ltd., De Trey Division, Weybridge, Surrey, U.K.; IRM®, L.D. Caulk Company, Dentsply International, Inc., Milford, DE 19963), and calcium hydroxide base materials (Dycal®, L.D. Caulk Company, Dentsply International, Inc., Milford, DE 19963), as well as a polycarboxylate cement (Poly

Received for publication August 22, 1985
Accepted for publication April 23, 1986

Fig. 1. — Low-power electron micrograph of a nitrocellulose replica. Arrows indicate the restoration margin (× 14). (RE) = replicated enamel; (RA) = replicated amalgam.
Ca being present in any of the seals analyzed. Finally, Sarkar et al. (1981) investigated the morphology and chemical nature of the corrosion products formed in an in vitro study of extracted teeth restored with different amalgam alloys and immersed in 1% NaCl at 37°C for nine months. Of note is the fact that sulfur was absent in all the marginal seals examined, even though the tooth itself is a source of sulfur (Jenkins, 1978). Nevertheless, despite the differences, all three reports concur in that Sn was found in large quantities at the amalgam-tooth interface. This is not surprising, since it has been established that the Sn-containing phase in amalgam is the most corrosion-prone. The high levels of Sn found in the seals of the present study once again confirm this.

In the two cases in this study where it was possible to analyze the marginal seal at both the enamel and dentin margin in the same specimen, Sn was present in greater amount at the dentin-amalgam interface than in the enamel interface. It is thought that the increased dentinal fluid flow in this area would enhance amalgam corrosion, and hence the migration of Sn. However, since only two samples were analyzed, this must be regarded as speculative at this stage.

The majority of Ca and P detected in the seal is probably derived from the base material and/or the tooth. Since all ten restoration bases analyzed contained P and Ca, it is likely that the bases served as a ready source of these two elements. Furthermore, since Marek et al. (1973) and Marek and Hochman (1975) have demonstrated that the interface crevice has a low pH, a demineralization of the tooth material may occur at this junction; Ca and P would then be available for seal formation.

The chlorine found in the seal probably came from the diet, saliva, and base materials. Since Copalite®, one of the controls, was shown to contain Cl, varnishes may also be a source for this element at the interface. Unfortunately, using the retrospective approach adopted here, it is impossible to assess whether a varnish had been used during cavity preparation. The role of varnishes in seal development needs to be clarified. Most of the relatively high levels of Zn found at the tooth-amalgam interface are likely to be from the base material. The passage of ions and/or elements from base materials to the interface is further supported by the detection of Al at the interface in specimens which were shown to have this element present in their base. Furthermore, two of the three specimens without detectable Zn at the interface also contained no Zn in their amalgams and base materials. Although the effects of bases on marginal seal formation and restoration longevity remain to be elucidated (Jodaikin, 1981), the numbers of specimens containing Zn in the seals in this study suggest that bases could play an active role in seal development.

The diet, saliva, and tooth, as such (Jenkins, 1978), are the probable sources of S, the amount of which varied among specimens of marginal seals. The greater quantity of this element at the amalgam-tooth interface in the two specimens where it was possible to analyze seal material at both the dentin and enamel interface further supports this view. It was notable that Cl was absent in the one specimen where S occurred as the predominant element in the seal material.

The two high-Cu (<10%) and one of the two intermediate (3-6%) Cu amalgams had Cu present in their marginal seals. Only three of the low-Cu (>2%) specimens were shown to have Cu present in their marginal seals. This is consistent with the finding that Cu has been detected in dentin as a corrosion product from low-Cu amalgams (Hals, 1976). It is not clear whether low-Cu amalgams release Cu into the tooth-amalgam interface or whether this is of clinical relevance.

The Fe detected in some amalgams and seals could have been derived from bleeding, from operative instruments, or

**Results.**

Visual examination of the fractured specimens showed that the only obvious differences, apart from the size of the restoration, were the thickness of the base material layer and its distribution on the cavity floor. When the nitrocellulose replica was examined in the scanning electron microscope, the marginal seal occurred as a continuous layer or as powder-like clumps along the well-replicated tooth-amalgam interfaces (Figs. 1 and 2). The results of the elemental analyses of the ten amalgams, base materials, and seals of the specimens are summarized in the Table. X-ray analyses of all the teeth showed that they were composed largely of Ca and P, but trace amounts of Na, Cl, and Mg were detected.

**Discussion.**

The patterns of elements found in this investigation are, in general, similar to those reported in similar studies (Wei and Ingram, 1969; Holland and Asgar, 1974; Sarkar et al., 1981). However, the methods and results of these latter investigations varied slightly from those of the present study. Wei and Ingram (1969) confined their analysis to Sn, Ag, and Hg at the amalgam-tooth interface of six silver-amalgam-restored molars which had been embedded in epoxy resin, sectioned, and polished. Holland and Asgar (1974) did a complete analysis of all elements present in the marginal seals of 12 sectioned, aged amalgam-restored teeth and stressed that these restorations were all in various degrees of deterioration. They made no mention of
TABLE

PRESENCE OF VARIOUS ELEMENTS WITHIN THE AMALGAM, BASE, AND SEAL OF TEN AMALGAM-RESTORED TEETH

| Specimen | Low Copper | | | | | Interm. Copper | | | | | High-copper |
|----------|------------|---|---|---|---|---|---|---|---|---|---|---|
|          | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
| Amalgam  | Ag | ++ | + | + | + | + | + | + | + | + | + | + |
|          | Cu | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ |
|          | Fe | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ |
|          | Hg | ++ | ++ | ++ | ++ | ++ | ++ | ++ | ++ | ++ | ++ | ++ |
|          | Sn | + | + | + | + | + | + | + | + | + | + | + |
| Other elements | P | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ |
| Base     | Ca | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ |
|          | Mg | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ |
|          | P  | + | + | + | + | + | + | + | + | + | + | + |
|          | Sn | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ |
|          | Zn | ++ | ++ | ++ | ++ | ++ | ++ | ++ | ++ | ++ | ++ | ++ |
| Other elements | Al~S~ | K~ | Al~ | Al+K~ | Fe~ | Cu~Fe~ | Al~S~ | Al~ | Al~Ba~ | S+ |
| Seal     | Ca | ++ | + | + | + | + | + | + | + | + | + | + |
|          | Cl | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ |
|          | Cu | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ |
|          | Fe | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ | ~ |
|          | P  | ++ | ++ | ++ | ++ | ++ | ++ | ++ | ++ | ++ | ++ | ++ |
|          | Sn | + | + | + | + | + | + | + | + | + | + | + |
|          | Zn | + | + | + | + | + | + | + | + | + | + | + |
| Other elements | Ag~ | Ag~ | Ag~ | Ag~ | Ag~ | Al~ | Al~ | Al~ | Al~ | Al~ | Al~ | Al~ |
| Key:     | ~ | element weakly represented      | + | element present        | + + | element strongly represented | D | dentin | E | enamel |

from the diet; it is unlikely to have originated from the saw, since the surfaces analyzed were fractured, not cut. The rôle of Fe in seal formation is unknown.

Sarkar et al. (1981) found Hg in various amounts on all corrosion surfaces examined. In the present investigation, Hg was absent from the seal material embedded in the replicas. This supports the opinion of Sarkar et al. (1981) that the Hg that they detected in seal material was generated from the underlying amalgam. The technique of using replicas to remove material from the amalgam-tooth interface for subsequent x-ray micro-analysis permits the visualization and accurate qualitative elemental analysis of the marginal seal in the same specimen, and lends itself to retrospective studies which may be correlated with the clinical performance of restorations and can provide a more accurate elemental composition (unpublished results).

This study emphasizes that marginal seal development is a dynamic process involving different elements from a variety of sources. The mechanism of marginal seal build-up needs to be investigated further so that the optimal conditions for its speedy and durable formation are understood. In the future, these conditions can then be created by clinical methods during cavity preparation, thus promoting restoration longevity.

Acknowledgments.

The authors would like to thank Professor P. Cleaton-Jones, Director of the Dental Research Institute, for his constructive criticism during the writing of this manuscript, and Mrs. H.A. Ball for the typing thereof.

REFERENCES