Analyses of the Amalgam-Tooth Interface Using the Electron Microprobe

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The electron microprobe was used to study tooth discoloration caused by old silver amalgam fillings. Oscilloscopic displays were obtained for the distribution of tin, silver, and mercury. In addition, a linear graphic display technic on the Enhancitron was used. The oscilloscopic displays showed the migration of tin from the amalgam into the enamel and dentin as opposed to silver or mercury, or both. These findings confirm that tooth discoloration is due to the presence of tin from the process of corrosion rather than from mercury.

Silver amalgam remains one of the most widely used filling materials in restorative dentistry. However, it becomes tarnished in the oral environment and tends to discolor the tooth substance. This discoloration increases with age and is most prominent at the amalgam-tooth interface.[1–3]

The nature of discoloration of tooth substance by silver amalgam has been studied by only a few workers.[4–6] The most widely accepted explanation for this discoloration is that metallic elements, principally mercury, penetrated into the enamel and dentin as a result of galvanic action.[6–8] Other studies,[5,9] however, showed that amalgam-discolored dentin contained mainly tin, with traces of silver, copper, and zinc, but no mercury.

The present study, therefore, used the electron microprobe to examine several interfaces between silver amalgam and enamel and dentin. The purpose was to determine which of the chemical elements migrate from the amalgam into the tooth substance and may be responsible for the tooth-discoloration phenomenon.

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Materials and Methods

ELECTRON MICROPROBE.—Microprobe analyses were carried out on an Applied Research Laboratory Electron Microprobe X-ray Analyser.* The semiquantitative display of the distribution of the elements studied was recorded on the oscilloscope and photographed by an attached Polaroid camera. In addition, a graphic display of the concentration of each element was obtained, using a linear graphic display technic on the Enhancitron.† The Enhancitron is a 1,026-channel data storage device which constructs a graph of the X-ray intensity over a line scan. In order to compare the distribution of elements both in enamel and in dentin, areas of interface between the amalgam and the dentinoenamel junction (DEJ) were selected for microprobe scanning. The main elements scanned were tin, silver, and mercury. The probe was operated at 25 kv and 0.05 μA for all scans, and the average spot size was approximately 1 μ. As the exact compositions of the old amalgams were unknown, no scanning was made for the detection of copper and zinc.

SAMPLE PREPARATION.—Six freshly extracted molars with large silver amalgam restorations which showed discoloration of the enamel and dentin were selected. The roots were removed, and the crowns were dehydrated through serial alcohol and were then embedded in a hard mix of epon. Embedded specimens were sectioned longitudinally with a diamond wheel in a thin sectioning machine. The specimens were polished, using fine abrasive paper down to grade 4/0 emery paper,‡ and then polished

* ARL, Sunland, Calif.

† Nuclear Data, Schaumburg, Ill.

‡ Buehler, Ltd., Evanston, Ill.

on metallurgical wheels, using fine-grain silicon carbide and aluminum oxide. Carbon was evaporated onto the sample surface, using a carbon evaporator,* before placement in the electron microprobe.

Results

Figure 1 shows the electron-probe scanning image of the tin distribution in an amalgam-tooth interfacial area. In all the scans, tin was rather unevenly distributed, and large numbers of "hot spots," or localized areas of high concentration of tin, were seen in the amalgam. Furthermore, there was a high concentration at the interface of the amalgam with the tooth surface. When a tracing of the amalgam-tooth interface was superimposed on this display, it was found that tin had migrated across and into the interfacial area as well as into the tooth substance. The amount of tin present in the dentin usually exceeded that in the enamel. A finger-like projection appeared to be present in the dentin just next to the DEJ, indicating a localized high concentration of tin.

Figure 2 is a typical picture of the electron-probe image for silver and was taken at the same interfacial area as Figure 1. It showed the nonhomogeneous distribution of silver in the amalgam. The distribution of silver did not correspond to the tin distribution. In fact, areas which were void of silver were found to exhibit a high tin concentration. There was no apparent migration of silver into the enamel or dentin; nor was there a high concentration at the interfacial area, as was seen with tin.

Figure 3 is the display of mercury of the same interfacial area as shown in Figures

* MIKROS VE 10, Portland, Ore.

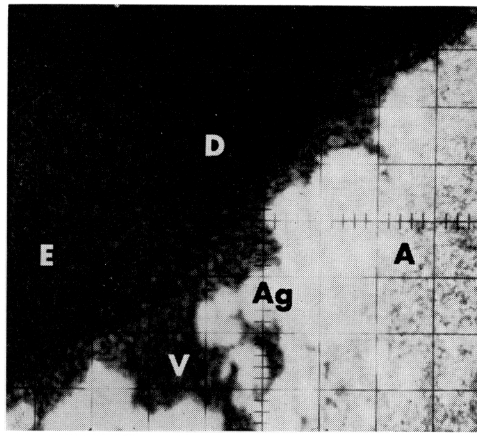

FIG 2.—Oscilloscopic display of the silver distribution (*Ag*) in silver amalgam (*A*) of the same interfacial area. An area void of silver is apparent (*V*). *E,* enamel; *D,* dentin (orig mag ×300).

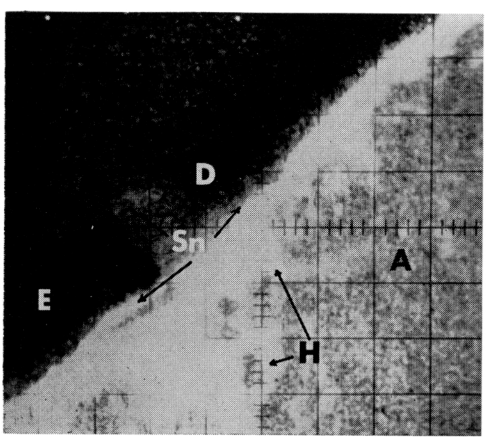

FIG 1.—Oscilloscopic display of the tin distribution (*Sn*) in silver amalgam (*A*) of an interfacial area. Note the localized high tin concentration (*H*) and the migration of tin across the interface into dentin (*D*). *E,* enamel (orig mag ×300).

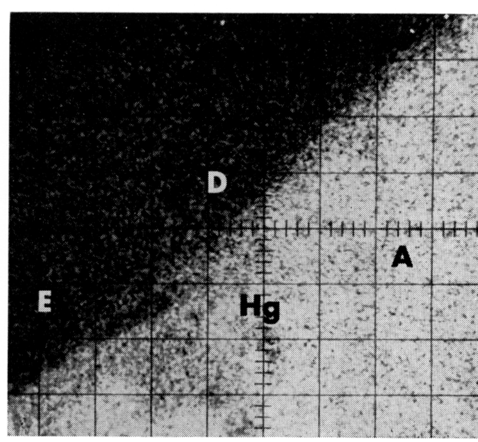

FIG 3.—Oscilloscopic display of the same interfacial area for mercury (Hg). Notice the more homogeneous distribution of mercury in silver amalgam (*A*). No migration of mercury into the tooth surface is observed. *E,* enamel; *D,* dentin (orig mag ×300).

1 and 2. In contrast to tin and silver, the distribution of mercury in amalgam was much more homogeneous. There was no apparent migration of mercury into the tooth surface or into the interface, and a sharp border between the amalgam and the tooth surface was usually observed.

Figures 4 and 5 are additional photographs for the distribution of tin and mercury, respectively. They confirm the migration of tin rather than mercury into the tooth substance.

Figure 6, *top,* shows the linear graphic display analyses of the penetration of tin into enamel. The undulating nature of the lower tracing indicated the nonhomogeneous distribution of tin. After crossing the inter-

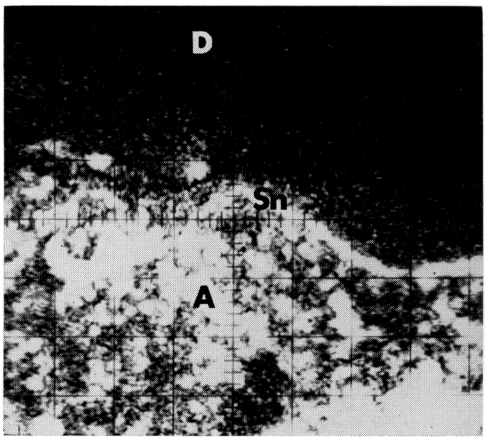

FIG 4.—Oscilloscopic display of another interfacial area showing the patchy distribution of tin and the migration of tin (*Sn*) into the dentin (*D*) from the silver amalgam (*A*) (orig mag ×300).

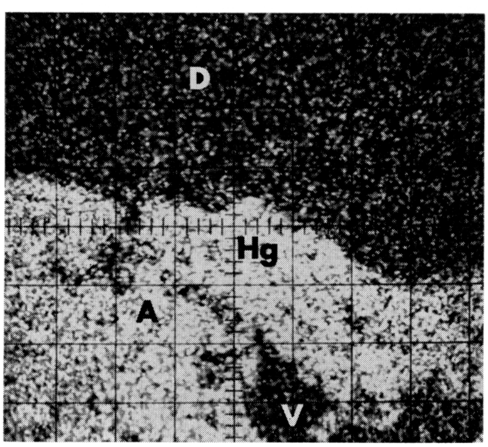

FIG 5.—An oscilloscopic display of the mercury distribution of the same interface as in Figure 4. No migration of mercury (*Hg*) into the tooth is seen. *A,* amalgam; *D,* dentin; *V,* void (orig mag ×300).

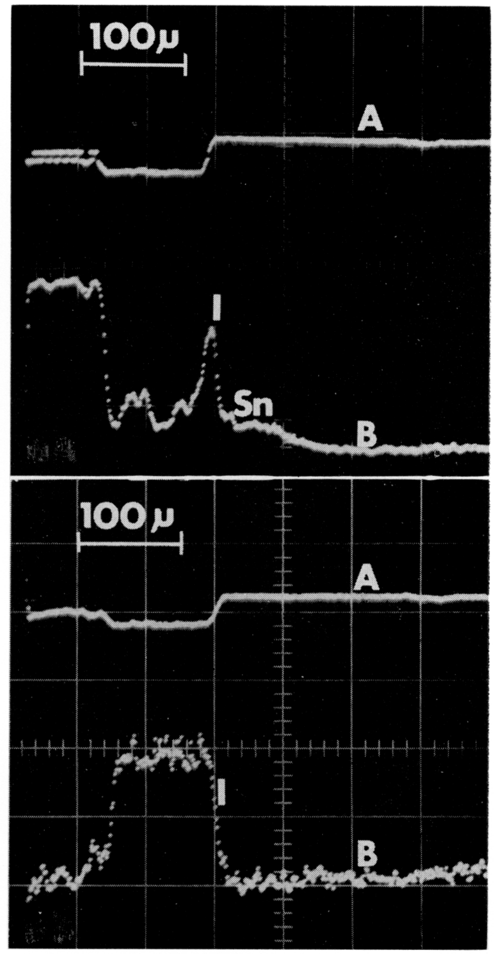

FIG 6.—The oscilloscopic displays of results obtained from the Enhancitron. The electron beam was scanned from the amalgam through the interfacial region (*I*) into the enamel. Line *A* shows the density of the specimen, and line *B* shows the concentration of the element in question along the path of the electron beam, plus the background intensity. Tin (*Sn*) is present after crossing the interface and extending into the enamel (*top*). The mercury concentration drops sharply at the interface, and only background intensity is recorded in the tooth substance (*bottom*).

face, a decreasing gradient of tin concentration was observed, and it was clear that tin was present in the immediate adjacent areas of the enamel.

Figure 6, *bottom,* shows the linear graphic display for mercury of the same area. No apparent migration of mercury from the amalgam to enamel had occurred, as evidenced by the abrupt drop in intensity of mercury at the interface.

Discussion

The electron-probe images showed qualitatively and semiquantitively structural nonhomogeneity of silver amalgam. Skinner and Phillips[7] suggested that the product of corrosion of silver amalgam in saliva is principally tin deposition with traces of silver and copper. The results of the present findings confirmed the foregoing theory.

A number of studies have used the electron microprobe to analyze the calcium and phosphorus concentrations in bone and dental tissues.[10–12] The present study points further to the applications of the electron microprobe to problems in dental research and especially to dental materials.

Conclusions

The electron microprobe was used to study the qualitative and semiquantitative distribution of silver, tin, and mercury at the amalgam-tooth interface. Oscilloscopic displays showed substantial migration of tin from the amalgam into the tooth substance. No mercury penetration was found. These results were confirmed by a linear graphic display technic on the Enhancitron.

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