Progress Report
on
CLINICAL INVESTIGATION OF A RADIOPAQUE
COMPOSITE RESTORATIVE MATERIAL
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NATIONAL BUREAU OF STANDARDS

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COMPOSITE RESTORATIVE MATERIAL

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This investigation was supported in part by Research Grants DE02494-02 and DE02742-01 to the American Dental Association from the National Institute of Dental Research and is part of the dental research program conducted by the National Bureau of Standards, in cooperation with the Council on Dental Research of the American Dental Association; the Dental Research Division of the United States Army Medical Research and Development Command; the Dental Sciences Division of the School of Aerospace Medicine, USAF; the National Institute of Dental Research; and the Veterans Administration.

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ABSTRACT

A radiopaque composite restorative material was developed using a ternary eutectic dimethacrylate liquid formulation as the binder and a BaF\textsubscript{2}-containing glass and fused silica as the reinforcing fillers. One hundred and ten restorations were placed using this newly developed material. The restorations will be observed over a prolonged period. They had sufficient radiopacity and yet were esthetically pleasing. Postoperative radiographs showed comparatively radiolucent areas in and around many of the restorations. These radiolucencies were usually caused by unfilled spaces at the tooth restoration interface.
The purpose of this paper is to report on the development of a radiopaque anterior restorative material, the initial esthetic properties of this material, and an investigation of the cause and prevention of one source of radiolucent areas observed in the postoperative radiographs of clinical restorations placed during this study.

One of the deficiencies of currently available esthetic anterior restorative materials is their radiolucency. Therefore, the dentist often cannot determine, from the radiographs alone, if a radiolucent area is an existing restoration or primary caries nor can he easily differentiate the restoration and recurrent caries. Neither can he judge if there is gingival overhang, sufficient contour, or complete filling of the preparation. It is also desirable to have all dental materials which are used in the mouth radiopaque so that if aspirated, ingested or inadvertently embedded in alveolar tissue they can be located radiographically.
Materials and methods

Since an esthetic restorative material must match the optical translucency of the natural teeth, it is necessary that the reinforcing fillers be transparent and that the finest particles (especially those less than several micrometers (microns) in size) have refractive indexes close to that of the polymeric binder. It is also essential to have a filler that can be chemically bonded to the resin by a suitable coupling agent. A radiopaque glass was formulated previously that filled these requirements. The glass used had the following batch formulation: SiO₂, 44; BaF₂, 28; B₂O₃, 16 and Al₂O₃, 12, in mole percent. The glass was ground to a very fine powder in a porcelain jar mill using quartz pebbles. The powder that passed through a U. S. Standard Sieve No. 400 [37 μm (micrometers; microns) and finer] was further classified into nearly equal parts of "coarser" and "finer" particles by sedimentation. The finer powder was used.

One part by weight of the BaF₂-containing glass was combined with two parts by weight of vitreous silica having
particles in the 5 to 37 µm size range. Although more (or less) BaF₂-containing glass may be used, it is desirable to use the least amount necessary for diagnostic purposes because it has a higher coefficient of thermal expansion [about 7.0 x 10⁻⁶/°C (28-62°C)] than does the fused silica [about 0.5 x 10⁻⁶/°C (25-50°C)] portion of the reinforcing fillers. The coefficient of thermal expansion of the resin binder is approximately 88 x 10⁻⁶/°C (26-48°C; dry).

The silica particles had been made spheroidal by a plasma device.⁵ Although it was not done, it is believed that it would be desirable to make spheres of the BaF₂-containing glass particles if a suitable refractive index could be maintained.⁴ The use of rounded reinforcing filler particles with a suitable size distribution results in an increased filler to resin ratio and thus a reduction in the coefficient of thermal expansion, an increase in stiffness, and in other desirable characteristics.³ The glass and fused silica powders were made hydrophobic by treatment with 0.5% 3-methacryloxypropyltrimethoxysilane; this treatment significantly improves the bonding between
the resin and the fillers. Benzoyl peroxide (0.50%) and lauroyl peroxide (0.82%) were dispersed into the mixed powders with the aid of a volatile solvent (ether). Seventy-nine parts by weight of this powder was mixed with 21 parts by weight of a ternary eutectic dimethacrylate liquid formulation that will be described elsewhere. The materials were mixed for one minute on a glass surface using an agate spatula.

One hundred and ten restorations (55 pairs) were placed clinically. In one of each pair the cavity surfaces were treated with an acetone solution of NPG-GMA[^6] [N-(2-hydroxy-3-methacryloxypropyl)-N-phenylglycine], an adhesion-promoting coupling agent.[^7] The other cavity preparation was treated with acetone alone as a control. The liquids were applied with a cotton pledget. Another cotton pledget was immediately used to remove the excess and to dry the preparation.

Small amounts of the composite material (having a thick putty-like consistency) were inserted into the preparation with a hand instrument. The instrument has a plugger at one end, 1.3 mm in diameter and a blade at the other end, about 2.0 mm long and 0.7 mm thick. With the plugger end an attempt
was made to pack the material into the retention forms that had been prepared with a No. 1/2 round bur. The preparation was then overfilled and grossly contoured with the blade end of the instrument. A matrix strip was then drawn tight and held motionless for two minutes beyond the time of the initial hardening of the material. All of the Class III restorations were placed using this technic which is a generally accepted method for the placement of silicate cement having a similar consistency.\textsuperscript{9,10}

The restorations were finished after a minimum of 24 hours using scalpels, stones, discs, and strips. The final surface was attained with fine cuttlefish strips or discs coated with a commercial lubricant often used for finishing silicate cement restorations. Postoperative radiographs were made at this time.

These paired restorations will be observed over a prolonged period to determine the effects of the use of the adhesion-promoting cavity primer (NPG-GMA) on changes in color, marginal integrity, and tooth sensitivity. The condition of adjacent soft tissues, changes in surface texture and contour, fractures, and recurrent caries will be noted. The foregoing will be followed by making gypsum
casts, color photographs, radiographs and by recorded visual and tactile inspection. The long-term clinical success of the formulation reported here remains to be ascertained; the results of these observations are planned for future publication.

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Results and discussion

After the restorations were finished their esthetic values were excellent and compared favorably with those of a commercially available radiolucent composite (Fig. 1).

Figure 2 shows four different radiographs of the same extracted tooth. The restoration has been replaced in each instance by one having a different amount of the BaF$_2$-containing glass as explained in the legend. All formulations appeared to be esthetically similar. The formulation for clinical evaluation (Fig. 2-B), given previously, was selected subjectively as having sufficient radiopacity for diagnostic purposes.

Figure 3 shows a radiograph of one of the radiopaque restorations. Radiolucent areas can be seen at the tooth-
restoration interface. Some form of radiolucency was observed around most of the radiopaque restorations that were evaluated radiographically.

To ascertain the causes of the radiolucent areas, restorations were placed in extracted teeth, radiographed and serially ground in about 10 increments per tooth. After each grinding, the tooth restoration interface was examined with the unaided eye and with an optical microscope and a correlation was made with the radiographic findings.

Figure 4-A pictures a radiograph of an extracted second molar with a finished mesial (1) and a finished distal restoration (2). Arrows point to radiolucent areas around the restorations. Both restorations were made using a technic similar to the one used clinically,9,10 except that the mesial restoration (1) was made by introducing a large excess of material into the preparation in one increment without packing or adapting it to the cavity walls with the hand instrument. The tooth was ground from the buccal surface in several increments and large unfilled spaces were uncovered. These spaces were filled with blue inlay wax for
contrast and photographs were taken (Fig. 4-B,C,D,E). Thus, a correlation was established between the radiolucencies in Figure 4-A and the voids shown in Figure 4-B,C,D,E.

The unfilled areas occurred most often in line angles, in retentive areas and in small preparations where access for the filling instrument was limited. These areas are also ones in which air could be trapped easily and thus could offer resistance to the flow of the material. It is evident in Figure 4 that the technic used in placing the mesial restoration (1) was inferior to that used in placing the distal restoration (2).

During the initial stages of the placement of the clinical restorations the operator became increasingly aware of the presence of the radiolucencies on the post-operative radiographs. Consequently, an attempt was made to avoid their occurrence by a more meticulous adaptation of the first increments of the material to the walls of the preparation. In subsequent radiographs there were fewer radiolucencies using the improved technic.
The results suggested that similar conditions exist in restorations made using other types of filling materials. An unusually critical examination of clinical radiographs of teeth containing silicate cement and resin restorations placed by many dentists was made, and similar radiolucencies were observed even though there was little contrast between the already translucent restorations and the underfilled areas. Similar radiolucencies could not be seen on radiographs of teeth containing amalgam restorations because the radiopacity of the material completely masked their presence. However, voids could be seen on serial grinding.

One could not be certain that all of the radiolucencies were caused by unfilled spaces or remaining decalcified dentin. Radiolucent borders or halo-like areas were frequently seen in radiographs of radiopaque composite restorations placed in non-carious extracted teeth in which no void spaces could be detected on serial grinding (Fig. 5). Thus, in some cases it was not known what caused the
radiolucencies; neither is it known if all the radiolucencies are of clinical importance unless they are misdiagnosed as caries.

Certainly all voids at the tooth-restoration interface are undesirable since they could serve as reservoirs where debris and bacteria could collect by percolation. The detection of these voids radiographically is one of the advantages of a radiopaque restorative material.

The experience gained in the development of this composite material is being used in efforts to make denture base materials radiopaque. Denture bases that can be located radiographically are needed in the event of accidental aspiration or ingestion.

Summary

Radiopaque esthetic direct filling materials are needed to facilitate dental radiographic diagnosis. A radiopaque composite restorative material was developed that has adequate esthetic properties. The resin binder is a ternary eutectic dimethacrylate formulation. The powder consists of a BaF₂-containing glass and fused silica both treated with a silane coupling agent and containing peroxide initiators.
One hundred and ten clinical restorations were placed using the above formulation. Post-operative radiographs showed radiolucent areas around many of the restorations. Sectioning of extracted teeth containing similar restorations revealed, in many instances, unfilled spaces which corresponded with the radiolucent areas on the films. These spaces are formed during the insertion of the plastic material into the cavity preparation and can be minimized in size and occurrence by careful adaptation of the initial increments of the restorative material to the surfaces of the preparation.

Such unfilled areas are not easily detected in radiographs of restorations that are either radiolucent or as radiopaque as amalgam.
References


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Legends

Figure 1: Photograph and radiograph of two Class III restorations made of composite materials. A is made from a commercial product and B from the experimental material. Both have good esthetic qualities but A is radiolucent while B is radiopaque.

Figure 2: Four different radiographs of the same extracted tooth. The restoration has been replaced in each instance by one having a different amount of the BaF₂-containing glass. A: 40% BaF₂-containing glass by weight; B: 26%; C: 20%, and D: 16%. Especially in C and D it is apparent that relative radiolucency occurs in areas (indicated by the arrows) where enamel is replaced by the somewhat less radiopaque composite material.
Figure 3: Radiograph of a clinical radiopaque restoration showing radiolucent areas designated by arrows.

Figure 4: A: Radiograph of an extracted tooth containing radiopaque composite restorations in the mesial (1) and distal (2) surfaces. Arrows designate radiolucent areas; B and C: Photographs of the tooth shown in A after grinding away the facial surface uncovering void areas in the mesial restoration. The void areas have been filled with blue inlay wax for contrast. The two sections (B and C) are about 1 mm apart; D and E: Same as B and C except distal restorations.

Figure 5: Radiograph of a non-carious extracted tooth containing mesial and distal radiopaque restorations. Note the radiolucent line at the tooth-restoration interfaces designated by arrows. No spaces could be detected when the tooth was serially ground.