A Comparative Study of Fluoride-releasing Adhesive Resin Materials

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Received September 25, 2001/Accepted January 17, 2002

One of the most important and exciting properties of recently introduced dental restorative materials is their ability to release fluoride ions, as this has several advantageous effects on tooth structures. They have been extensively used as fluoride-releasing filling and luting materials. Recently, fluoride-releasing adhesive resins and fluoride-releasing adhesive resin cement have been developed and introduced for clinical use. The purpose of this study was to evaluate the fluoride release from these adhesive resins and the fluoride uptake by both enamel and dentin, as well as the acid-resistance of these tooth structures. Based on our results, we conclude that fluoride-releasing adhesive resins and luting cements are useful for the prevention of initial or secondary caries, especially along the margins of restorations.

Key words : Fluoride release, Fluoride uptake, Secondary caries

INTRODUCTION

The cariostatic effect of resin-based pit and fissure sealants on the occlusal surfaces of primary and permanent molars has previously been reported¹⁾. The incidence and severity of secondary caries in tooth structures were found to have been reduced around fluoride-releasing materials^{2,3)}. Glass-ionomer cements have been traditionally used as pit and fissure sealants because of their fluoride-releasing and cariostatic effect on teeth. The traditional glass-ionomer cements were the product of an acid-base reaction between basic fluoroaluminosilicate glass powder and poly-carboxylic acid in the presence of water. The nature of the set cement comprised an organic-inorganic complex with high molecular weight. Glass-ionomer cement can, thus, be defined as a water-based material that hardens following an acid-base reaction between fluoroaluminosilicate glass powder and an aqueous solution of polyacid. As such, glass-ionomer cements are able to release fluoride⁴⁾. From the clinical aspect, the use of glass-ionomer cement as a permanent filling was limited, and was mainly used as a temporary restoration, fissure sealant or for partially erupted teeth where isolation from saliva is difficult and caries risk status demands immediate protection^{5,6)}.

Today, glass-ionomer cements are widely used in dentistry because of a variety of beneficial properties that include chemical diffusion-based adhesion to enamel and dentin, fluoride release, biocompatibility with tooth structure, simple application,

esthetic appearance, acceptable abrasion resistance and capacity to be retained on unsupported enamel⁷⁾. The two main reasons for the popularity of glass-ionomers are their permanent ionic bond to tooth structures, and their capacity to release fluoride, making them useful materials to replace dentin when used as a base in deep cavities^{8,9)}. Glass-ionomer cements have been recommended for use as base in conjunction with resin composites, and the so-called sandwich or laminate technique has become a common practical procedure to take advantage of glass-ionomer cement's fluoride release in combination with resin composites for restorations^{10,11)}. This so-called sandwich or laminate technique utilizes the fluoride-releasing glass-ionomer cement as a lining or base material, and composite resin for filling.

Presently, the composite resin restoration technique has become essential in dental clinics and the use of adhesive agents has become indispensable for composite resin restorations. However, this technique is very difficult for cavity filling, most especially when a perfect composite resin restoration is desired. As a result, various fluoride-releasing adhesive materials for composite resin restoratives have been developed and introduced with an inhibitory action against secondary caries arising from enamel cracks or micro-leakage on the tooth-restoration interface. It has been demonstrated that composite resin restorations occasionally exhibit enamel cracks (on the occlusion side) as well as micro-leakage (contraction gaps on the gingival side)^{12,13)}. These result mainly in the formation of secondary caries. Fortunately, these can be prevented by the action of fluoride ions released from fluoridated liners. This method of fluoride release from restorations is a clinically safe method for caries prevention because the presence of fluoride increases the acid resistance of the tooth structure that comes in contact with it, thus exerting a cariostatic effect¹⁴⁻¹⁹⁾.

The purpose of this study was to evaluate fluoride release from fluoridated adhesive materials, fluoride uptake by enamel and dentin fluoride from test materials as well as improvement in the acid resistance of tooth structures through fluoride-releasing resin bonding systems.

MATERIALS AND METHODS

The materials used in this experiment are indicated in Table 1. A Bis-GMA type bonding agent of traditional adhesive resin (Clearfil Photobond, Kuraray, Osaka, Japan) with no fluoride-containing ingredients was used as a control material.

Information on the fluoride content of the test materials was based on the data provided by the manufacturers.

The following investigations were conducted:

1. Fluoride release

Specimens were prepared using a plastic ring mould (with internal dimensions of 9 mm diameter and 1 mm height). The mould with test materials was held between two glass slides and covered with a transparent polyester strip. The materials were manipulated according to the manufacturers' instructions. A silk thread was placed

Table 1 Materials, manufacturers, batch numbers and system compositions

Material	Code	Material composition	Manufacturer	Batch No.
ABF	ABF	Primer: MDP, HEMA, Water, TEGDMA, PI, MDPB Bond: MDP, HEMA, TEGDMA, Microfiller, PI, Fluoride-Sodium	Kuraray, Osaka Japan	Experimental material
Photobond*	CPB	Conditioner: Phosphoric acid Bond: Bis-GMA, MDP, HEMA, TEGDMA, PI	Kuraray, Osaka Japan	11122
Fluorobond	FB	Primer: Water, catalyst HEMA, 4AET, 4-AETA, UDMA Bond: GI, UDMA, TEGDMA, HEMA, EGDMA, 4-AET, PI	Shofu, Tokyo Japan	PN1330
Fluorocement	FC	Primer: MDP, HEMA, 5-NMSA Cement: MDP, HEMA, TEGDMA, PI, Fluoride-Sodium	Kuraray, Osaka Japan	1080
One-Up Bond F	OBF	Primer & Bond: MDP, HEMA, Water, TEGDMA, PI Fluoride-silicate glass, methacrylate-10	Tokuyama Tokyo, Japan	45023

*: for Control

so that after the material set, it could be used to suspend the specimen in the solution. The beakers had tightly fitting lids to prevent evaporation of the solution. Specimens were immersed in deionized water (10 ml) at a temperature of 37°C (± 0.5). To analyze of fluoride ions, standards were prepared from sodium fluoride solution with concentrations of 0.1, 1, 10 & 100 ppm to which was added TISAB III (total ionic strength adjustor and buffer, Orion, Boston, USA) in order to obtain a constant background ionic strength.

Standard solutions were used to plot the calibration graph. Fluoride release was detected using a fluoride ion selective electrode (9609BN, Orion Model 290A, USA) connected to a microprocessor ion analyzer (Orion model 290A, USA). The test specimens were placed in a non-heating magnetic stirrer (SR 50, Advantec, Kanazawa, Japan) to obtain an even distribution of the fluoride ion. The temperature of the solution was adjusted to 23°C (± 2) in order to compensate for any fluctuation in temperature. The fluoride ion concentration (ppm) of the test solutions was recorded for all test specimens after immersion in deionized water for 1, 2, 3, 7, 28 and 56 days. Five specimens were prepared for each test material. Mean values of fluoride concentration were calculated each day and expressed in ppm. Data were analyzed by one-way analysis of variance (ANOVA) with the level of significance set at 5%.

2. Enamel and dentin fluoride uptake

The specimens were prepared from premolars that were extracted for orthodontic reasons, and stored at 4°C in isotonic saline no longer than 1 week before use. A class V cavity with a depth of about 2 mm was prepared on the cervical area of the enamel and dentin with a high-speed hand piece and round diamond burs. The cavity wall and floors were treated with the test material conditioner. The pre-treated cavity was restored with composite resin (APX, Kuraray, Osaka, Japan) and light cured

for 20-30 seconds using a visible light unit (LINHTEL-II, Kuraray, Osaka, Japan). One specimen was retained a polyethylene bottle (500 ml deionized water) that was changed with a fresh stock every 48 hours and maintained at a temperature of 37 °C after the margins were polished with a fine diamond point. The specimens were cross-sectioned longitudinally through the cavity center with a low speed diamond micro-cutter (MC 201, Micro cutter 201, Maruto, Tokyo, Japan) after being embedded in by epoxy resin (Quetol-812, Nissin EM, Tokyo, Japan) and polished with car-bide paper (No.600-1200) after immersion in deionized water for a 60-day period. Specimens were viewed with an electron probe X-ray microanalyzer WDX type (WDX, EPMA8705, Shimadzu, Kyoto, Japan) for elemental distribution of fluoride (F) on the adhesive materials-enamel or dentin interface. Five specimens were prepared for each test material.

3. Acid resistance

The materials and specimens used were prepared as above (fluoride uptake by tooth structure). The specimens were subsequently immersed in 20 ml acetic acid sodium buffer (acetic acid: 0.24 ml, sodium acetate, anhydrous: 0.33 g dissolved in 20 ml deionized water, and adjusted to 0.2 M, 4.5 pH) for 12 hours for demineralization in an incubator at 37°C (± 0.5) after storage at 37°C (± 0.5) for 60 days. The specimens were cross-sectioned longitudinally through the center of the restoration and were cut into several thin slices (60-80 μ m) with a low speed diamond micro-cutter (MC201, Maruto, Tokyo, Japan) after being embedded in epoxy resin (Quetol-812, Nissin EM, Tokyo, Japan) and polished by whetstone and observations of resin-enamel and dentin cervical marginal area of restorations were carried out with a Polarized Light Microscope (PLM). Five specimens were prepared for each test material.

Fig. 1 Accumulated fluoride released from fluoride adhesive resin materials.

Fig. 2

Fig. 3

Fig. 4

Fig. 5

- Fig. 2 Fluoride ion uptake by enamel and dentin on fluoridate adhesive resin materials ABF and *Fluorobond* (upper: 1 day; lower: 60 days after immersion in deionized water) ($\rightarrow$: rather high density of fluoride ion layer; E: enamel; D: dentin; R: resin; measurement bar : $20\mu\text{m}$).
- Fig. 3 Fluoride ion uptake by enamel and dentin on fluoridate adhesive resin materials *Fluorocement* and *One-up Bond F* (upper: 1 day; lower: 60 days after immersion in deionized water) ($\rightarrow$: rather high density of fluoride ion layer; E: enamel; D: dentin; R: resin; measurement bar: $20\mu\text{m}$).
- Fig. 4 Polarized light microscope picture of enamel and dentin samples after using ABF, *Fluorobond* and *Fluorocement*. (measurement bar: $100\mu\text{m}$).
- Fig. 5 Polarized light microscope picture of enamel and dentin samples after using *One-up Bond F* and *Photobond* (as control) (measurement bar: $100\mu\text{m}$).

RESULTS

1. Fluoride release

Patterns and the amount of cumulative fluoride release ($\mu\text{g}/\text{cm}^2$) from the specimens at days 1-56 are shown in Fig.1. The amount of cumulative fluoride released from the ABF and FC specimens when comparing the materials at first-day were statistically different from the FB or OBF. The amount of fluoride released at day 56 ranged from 2.9 to $28.1\mu\text{g}/\text{cm}^2$. The greatest was from FC ($28.1\mu\text{g}/\text{cm}^2$), followed by ABF ($10.5\mu\text{g}/\text{cm}^2$), OBF ($7.5\mu\text{g}/\text{cm}^2$) and FB ($4.4\mu\text{g}/\text{cm}^2$). There is a resemblance between the ABF, FB and OBF in the amount of fluoride released after the first day up to 60-day periods. FC showed the highest amount of fluoride release of all test materials over the whole test period. All test materials showed stability and a low level of fluoride release after the first day of the study period.

2. Enamel and dentin fluoride uptake

Figs. 2 and 3 show representative enamel and dentin specimens of the WDX result of contact interfaces around those test materials after 1 and 60-day periods. WDX analysis figures showed higher fluoride ion density in dentin and enamel contact areas, especially in the 60-day specimens when compared with 1-day specimens immersed in deionized water. Figs. 2 and 3 also showed that the fluoride density was higher in the dentin, when compared with that in the enamel contact area. In enamel and dentin fluoride was observed to depths ranging from $5\text{-}30\mu\text{m}$ from the contact surface. In FC, the fluoride uptake in the dentin contact interface was the deepest ($30\mu\text{m}$) of all the test materials.

3. Effect in acid resistance

Polarized light micrograph images in Figs. 4, 5 show artificial cervical surface secondary caries formation on a restored cavity with test materials after demineralization using acetic acid sodium buffer. This resulted in an almost acid resistant area in all the test fluoride-releasing materials in comparison with control specimens (Fig. 5 Photobond as control).

Fluoride release resin material restoration specimens in each case showed that

the acid resistance zone of the test materials was larger than in the control specimen (Photobond) in both enamel and dentin. The control specimen showed an outer lesion around the restoration compared with the fluoride release material specimens. The control specimen showed outer lesions ranging from 100-150 μm in depth, and about 20 μm wall lesion in restoration contact margins.

DISCUSSION

Many investigators have demonstrated the possibility of increasing fluoride ion density in enamel and dentin adjacent to a restoration when fluoride-releasing dental materials are used²⁰⁻²⁵. This resulted in an increased acid resistance in the margins of enamel and dentin in contact with fluoride-releasing materials thus exerting a cariostatic effect. Consequently, the use of fluoride ion releasing materials in restoring cavities has become an important approach in operative dentistry to prevent the development of secondary caries beneath restorations.

Investigators have demonstrated various levels of fluoride release and caries inhibition with these improved materials¹⁵. Recently, resin composites have been selected as restorative materials and the release of fluoride from these materials has been evaluated^{16,23}. Manufacturers have tried to develop a true fluoride-releasing composite resin with long-term resistance to after its condensation in prepared cavities. As a result, various fluoride-releasing adhesion systems and composite resins have been developed for clinical dental practice. Of special interest is the development of fluoride-releasing materials in glass-ionomer fillers (GIF, gel condition) or sodium fluoride (NaF) fillers added to the base resin of the adhesive materials. These new types of fluoride-releasing materials allow a more efficient release of fluoride ions as compared with a fluoridated polymer resin-modified sealant or resin composite (called compomer). The findings of this study clearly showed that fluoride release was greater in ABF and FC which contained GIF (fluoride-sodium fillers) materials as compared with FB and OBF that contained NaF (glass monomer gel fillers) materials during the 7-day period. However, the total amount of fluoride released in all the test materials was the same after a 7-day period, suggesting that the long-term fluoride release pattern in all test materials were essentially equal. This could be due to the presence of some sodium fluoride (NaF) fillers that make up fluoridated adhesive resin materials (ABF and FC). The release of fluoride ions from these materials seems to be more efficient compared with glass-ionomer gel fillers in fluoride-releasing adhesive resin materials (FB and OBF). The characteristics of fluoride-sodium fillers and glass-ionomer gel fillers are unknown and could not be revealed by the manufacturers but it is already known that the fluoride-sodium filler is a pre-treated inorganic compound which breaks into pieces, a characteristic of sodium that makes it easily soluble in water. Therefore, this could possibly explain the efficient release of fluoride ions contained in fluoride-sodium filler type fluoridated adhesive resin.

Many studies have demonstrated the ability of glass-ionomer cement restorations

in prepared cavities to increase fluoride content in the adjacent enamel^{26,27}. In this study, areas of enamel and dentin adjacent to fluoridated adhesive resin restorative materials exhibited rather high readings of the fluoride ion density in the enamel and dentin. The characteristic high density of fluoride ion layer was higher in dentin when compared with enamel. The density of the fluoride ion layer was greater over a 60-day period than a 1-day period in all the test materials. Fluoride uptake by dentin was easier due to the following properties of dentin over enamel, namely: a) the presence of moist dentinal tubules on the dentin surface, b) a well-moistened environment due to the presence of a collagen fiber network, c) dentin apatite crystals are smaller in size compared with enamel (enamel crystal length: 3000-5000 Å and width: 500-1200 Å; dentin crystal length: 200-300 Å and width: 40-70 Å) and lower in density, d) dentin contains more water compared with enamel (enamel is practically a dry structure).

The ability of enamel and dentin to absorb fluoride ions is an important property of these tissues. Moisture is an important media for fluoride ion release and uptake by tooth structure. Enamel is a tissue consisting mostly of inorganic matter; it is a dry tissue compared with dentin. Thus, the uptake of fluoride ion by enamel is more difficult than in dentin. Figs. 2 and 3 show a deeper and higher density of fluoride ion uptake by enamel and dentin. Fluoride ion uptake was greater for those containing fluoride-sodium filler materials than those with glass-ionomer gel fillers materials.

Recurrent caries or secondary caries are recognized as major causes of the failure of composite restorations²⁸⁻³⁰. This is probably due to the insufficient marginal adaptation of composite restorations, resulting in micro-leakage between the filling and hard tissue. All composite restoration walls prepared *in vitro* show some micro-leakage as estimated by dye penetration³¹. Generally, micro-leakage occurs between etched enamel and dentin restoration and is particularly extended at the cervical cavity wall below the enamel-cement junction²⁸⁻³⁰. Observable marginal adaptation defects have been reported in 50% of the hybrid composites and in 44% of the micro-filler resin restorations after 5 years³².

Furthermore, Wilson *et al.*³² presented evidence that 48% of all posterior composite restorations studies had a deteriorated marginal adaptation after 4 years. In contrast, 90% of composite resin restorations studied were still clinically acceptable after 5 years³³.

Composite resin restorations do show problems in due time as a result of micro-leakage and insufficient marginal adaptation. Therefore, fluoride-releasing restorative materials are worthy of investigation as they are claimed to possess recurrent caries-inhibiting properties. Recently, it has been shown that fluoride-releasing composite restorations reduced the lesion depth and mineral loss of adjacent enamel by about 30% *in vitro* studies³⁴.

Figs. 4 and 5 show the acid resistance phenomenon on enamel and dentin around fluoridated material restoration when compared with control specimens, as shown in Fig. 5 (as Photobond). ABF specimens did not exhibit caries-like lesions in the

enamel and dentin-resin interfaces and showed small outer marginal lesions (outer lesion) of about $30\mu\text{m}$ that compare well with the control specimen (about $100\mu\text{m}$). Other specimens did not exhibit wall lesions and/or outer lesions. An imperfect demineralization phenomenon was shown in FB and OBF, indicating that fluoride ion uptake by enamel and dentin inhibited the effect of acids.

There is a high possibility of the presence of caries around the margins of restorations; the application of fluoridated adhesive resins will decrease this possibility and inhibit secondary or initial caries around the margins of restorations in both enamel and dentin³⁵⁻³⁷.

The study must be tempered with the knowledge that the mechanism of fluoride action in caries reduction is not fully understood for the clinical implications, the optimum fluoride concentration for caries inhibition is unknown, the mechanism of decrease of secondary decay around restoration of the fluoride-releasing materials and clinical studies on fluoridated materials are lacking. However, it is worth noting that fluoride-releasing composites appear to have a local positive effect on the demineralization process of enamel and dentin, and may have a role to play in the future prevention of secondary caries³⁸.

CONCLUSIONS

In this study, fluoride ions released from fluoridated adhesive resin and luting cement was confirmed. Fluoride ion uptake by enamel and dentin was also observed to increase with the subsequent acid resistance of hard tooth tissues. The new fluoride-releasing restorative materials may prevent the development of secondary carious lesions and initiation of the carious process in the adjacent tooth tissue. This suggests the promise of caries inhibition when using the fluoridated materials clinically. We conclude that fluoride-releasing bonding resin and luting cement are useful to prevent initiation of the carious process and the development of secondary caries in restored teeth.

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