Evaluation of a New Fluoride-releasing One-step Adhesive

Linlin HAN, Akira OKAMOTO, Masayoshi FUKUSHIMA and Takashi OKIJI
Division of Cariology (Operative Dentistry and Endodontics), Department of Oral Health Science, Course for Oral Life Science, Graduate School of Medical and Dental Sciences, Niigata University, 5274 Gakkocho-dori 2-bancho, Niigata 951-8161 Japan
Corresponding author, Linlin Han E-mail: han4378@yahoo.co.jp

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In this study, a newly developed S-PRG (surface pre-reacted glass-ionomer) filler-containing one-step adhesive, called SI (SI-IB551, Prototype), was evaluated regarding its fluoride release behavior. As a result, fluoride ion distribution in the tooth structures and acid resistance of cavity margins were also evaluated. In addition, Absolute® (a fluoride-releasing one-step adhesive; AB) was evaluated in comparison to G-Bond® (a non-fluoride-releasing one-step adhesive; GB) which was used as a negative control. Concentration of fluoride released was measured using a fluoride ion selective electrode after immersion of each material in distilled water. Fluoride distribution in tooth-adhesive interfacial areas was examined with EPMA following cavity preparation in human premolars and having treated resin restorations with each material. To evaluate acid resistance, restored specimens were immersed in an acetic acid buffer (0.2 M, pH 4.5) for 12 hours and then the cavity margins were observed using scanning electron microscopy. Amounts of fluoride released from AB and SI were significantly greater than that from GB. Further, significant differences in fluoride release were detected between AB and SI. A layer of increased fluoride density was clearly detected at the enamel- and dentin-SI interfaces. In terms of acid resistance, an acid resistance zone was also formed adjacent to the tooth-adhesive interface of AB and SI specimens. However, in GB specimen, acid resistance zone was not observed. These findings suggested that one-step adhesives displayed a favorable fluoride release property, thereby contributing positively to inhibition of recurrent caries.

Key words : One-step adhesive, Fluoride-releasing, S-PRG filler

INTRODUCTION

The incidence and severity of secondary caries has been demonstrated to decrease around fluoride-releasing materials1–6). The use of composite resin restoration techniques has recently become an essential procedure in clinical dentistry, although it is not always possible to obtain a perfect marginal seal around the restoration. To overcome this problem, fluoride-releasing adhesive materials for composite resin restoratives have been introduced7).

Fluoride-releasing adhesives are expected to show an inhibitory action against secondary caries, because the fluoride released should help to reduce the demineralization caused by enamel cracks or microleakage from the tooth-restoration interface6–8). It is noteworthy that many in vivo and in vitro studies have reported that fluoride-releasing restorative materials rendered the resin-enamel and resin-dentin interfaces more acid-resistant, thereby exerting a cariostatic effect9–13).

Recently, a new type of self-etching agent, S-PRG (surface pre-reacted glass-ionomer) filler-containing one-step adhesive (and thus fluoride-containing) has been developed. Most importantly, it has received considerable acceptance due its simplified application procedure. Apart from this advantage, these products are expected to contribute effectively to inhibiting recurrent caries because being in direct contact with the tooth structure, they can act as a proximate reservoir of fluoride ions for uptake by enamel and dentin. However, the cariostatic activity of the newly developed adhesive is yet to be fully investigated.

Therefore, the purpose of this in vitro study was to evaluate the S-PRG filler-containing one-step adhesive in terms of its fluoride release. In this connection, the uptake of released fluoride, as well as the acid resistance, of both enamel and dentin were also examined.

MATERIALS AND METHODS

The fluoride-releasing and control one-step adhesives used in the study are indicated in Table 1. A non-fluoride-containing flowable composite resin FF (Filttek® Flow, 3M, MN, USA) was used as the cavity filling material. The following investigations were then conducted.

Fluoride release

Specimens were prepared using a plastic ring mold with an internal diameter of 9 mm and height of 1 mm. The molds with test materials were held between two glass slides and then covered with a transparent polyester strip. The materials were manipulated according to the manufacturers’ instructions, and five specimens were prepared for each test
material. Specimens were then kept in distilled water (10 ml) at a temperature of 37°C in an incubator (MIR-162, Sanyo, Osaka, Japan), using beakers with tightly fitting lids to prevent solution evaporation. Storage medium was collected at 1, 3, 7, 14, 30, and 60 days, and a fresh solution was then supplied. The medium was also replaced 24 hours prior to the collection, so that the amount of fluoride that had been released for 24 hours could thus be determined.

Fluoride concentration in the medium was determined using a fluoride ion selective electrode (9609BN, Orion, Boston, USA) connected to a microprocessor ion analyzer (model 290A, Orion, Boston, USA). The electrode was previously calibrated with fluoride standard solutions (0.1, 1, 10, and 100 ppm). Five milliliters of each test solution was then added with a magnetic stirrer and buffer; Orion, Boston, USA, without heating. Temperature of the solution was adjusted to 23°C to compensate for any fluctuation in temperature. Fluoride ion concentration (ppm) of each test solution was then measured. The data were expressed as the mean amount of fluoride per unit area of the specimen (μg/cm²). Differences in fluoride release were compared and tested for statistical significance by the t-test for independent variables. The significance level adopted for all tests was conventionally established at 0.05. Statistical analysis was performed using a statistical software package, SPSS (SPSS 11.0J, SPSS Japan Inc., Tokyo, Japan).

Fluoride uptake by enamel and dentin

All specimens were prepared from premolars that were extracted for orthodontic reasons and stored at 4°C in isotonic saline not exceeding three months. A cavity with a depth of about 2 mm and a mesiodistal width of about 4 mm was prepared in the cervical area (with cervical margins located in the root dentin) by means of a high-speed handpiece and round diamond burs (F440, Shofu, Kyoto, Japan). The cavities were treated with the test and control adhesives (SI, AB, and GB), filled with the composite resin FF, and light-cured for 30 seconds using a visible light unit (XL3000, 3M, MN, USA). Margins of the restoration were then polished with a fine diamond point (C22ff, Shofu, Kyoto, Japan).

Following storage in 500 ml of distilled water (replaced every 48 hours) at 37°C for 7 and 60 days, the specimens were then cross-sectioned longitudinally through the center of the cavity with a low speed diamond microcutter (MC 201, Maruto, Tokyo, Japan). They were then analyzed with a WDX type electron probe X-ray microanalyzer (EPMA; EPMA8705, Shimadzu, Kyoto, Japan) for the elemental distribution of fluoride in the enamel- and dentin-adhesive interfacial areas.

Acid resistance

All specimens were prepared in the same way as mentioned above (i.e., Fluoride uptake by enamel and dentin).

After storage at 37°C for 60 days, the specimens were stored in 100 ml of an acetic acid-sodium buffer (0.2 mol/L, pH 4.5) for 12 hours with stirring. They were then cross-sectioned longitudinally through the center of the restoration and cut into two pieces with the low-speed diamond microcutter. Enamel and dentin cavity margins were observed using a scanning electron microscope (SEM; HITACHI, S430, Tokyo, Japan).

Table 1 Adhesive materials and composite resin used

<table>
<thead>
<tr>
<th>Material</th>
<th>Code</th>
<th>Application</th>
<th>Composition</th>
<th>Type</th>
<th>Lot No.</th>
<th>Manufacturer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Absolute1</td>
<td>AB</td>
<td>Wet technique: 5 sec /2 times, LC: 10 sec.</td>
<td>Methacrylate, phosphoric acid, acetone, PEM-F</td>
<td>One-bottle</td>
<td>387-003</td>
<td>Dentply-Sankin, Tokyo, Japan</td>
</tr>
<tr>
<td>G-Bond</td>
<td>GB</td>
<td>Apply: 10 sec. LC: 10 sec.</td>
<td>Methacrylate, phosphoric acid esters, 4-MET, UDMA, acetone, water, Si</td>
<td>One-bottle</td>
<td>GB191</td>
<td>GC, Tokyo, Japan</td>
</tr>
<tr>
<td>SI-IB 5512</td>
<td>SI</td>
<td>Mixing: &lt;1 min. Apply: 20 sec. LC: 10 sec.</td>
<td>6-MHPA, 4-AET, 4-AETA, FASG, Bis-GMA, HEMA, S-PRG</td>
<td>Two-bottle</td>
<td>EXP</td>
<td>Shofu, Tokyo, Japan</td>
</tr>
<tr>
<td>Filtek Flow</td>
<td>FF</td>
<td></td>
<td>Bis-GMA, TEGDMA, UDMA, Titanium dioxide</td>
<td>Flowable resin</td>
<td>3BF</td>
<td>3M, ESPE, MN, USA</td>
</tr>
</tbody>
</table>

Material information as obtained from manufacturers:
1: Fluoride-containing adhesive (contains Pentamethacryloxyethyl cycophosphazene monofluoride, PEM-F);
2: Fluoride-containing adhesive (contains Surface Pre-reacted Glass-ionomer filler, S-PRG filler).
RESULTS

**Fluoride release**

Fig. 1 shows the amount of fluoride released from each material at each time point for a total observation period of 60 days. Fluoride release was detected from AB and SI, whereas none was detected from GB. In particular, fluoride release was greater in AB than in SI. Significant differences in fluoride release rate for AB, GB, and SI are also shown in Fig. 1.

**Fluoride uptake by enamel and dentin**

Figs. 2 and 3 show the results of a WDX element analysis on the interfacial areas of representative specimens (after storing in distilled water for 7 and 60 days, respectively). The SI specimens clearly showed an increased fluoride ion density in dentin ranging from 2 μm (Fig. 2) to 8 μm (Fig. 3) after being storing in distilled water for 7 and 60 days, respectively. Moreover, the fluoride ion density was higher in the 60-day specimen than in the 7-day specimen. However, the SI-enamel specimens failed to clearly show an increased fluoride ion density (for both 7 and 60 days).

In contrast, AB specimens showed a very low level of fluoride ion density in both enamel and dentin after 7 and 60 days of storage in distilled water. With GB specimens, there was no detectable fluoride uptake by enamel and dentin (Figs. 2 and 3).
Fig. 3  Fluoride uptake by enamel and dentin after storage in distilled water for 60 days (upper: enamel; lower: dentin).

Fig. 4  SEM pictures of AB, GB, and SI adhesive specimens. Acid resistance zone is formed in the dentin adjacent to fluoride-releasing adhesives (AB and SI). However, wall lesion and outer lesion are seen in the dentin adjacent to non-fluoride-releasing adhesive GB.
Acid resistance
Fig. 4 shows the SEM photographs of the dentin cavity margin of representative dentin specimens after demineralization with the acetic acid-sodium buffer. SI and AB specimens showed the formation of a demineralization inhibition zone adjacent to the dentin cavity margin. The demineralization inhibition zone was also observed at the enamel margin of SI and AB specimens (Figure is omitted). However, GB specimens showed highly demineralized zones (wall lesion and outer lesion) in both dentin and enamel (Figure is omitted).

DISCUSSION
Recurrent or secondary caries is recognized as a major cause for the replacement of composite restorations. In many cases, this may be caused by an insufficient marginal adaptation of composite restorations, which may lead to the occurrence of microleakage between the restoration and tooth structure. Cavity walls of composite restorations at various anatomical locations—prepared in vitro—have been demonstrated to show some degree of microleakage as estimated by dye penetration. In general, microleakage frequently occurs at the etched dentin-restoration interface and particularly extends at the cervical cavity wall below the dentin-enamel junction. Clinically, observable marginal adaptation defects have been reported to occur in 50% of hybrid composites and 44% of microfiller resin restorations after five years. Besides, Wilson et al. also presented evidence that 48% of all posterior composite restorations studied showed a deteriorated marginal adaptation after four years. Fluoride-releasing restorative materials are thus considered to be worthy of investigation as they are claimed to possess recurrent caries-inhibiting properties. Indeed, an in vitro study showed that fluoride-releasing restoration materials reduced the lesion depth and mineral loss of adjacent enamel by about 30%.

Apart from Forsten, many investigators have shown the ability of fluoride-releasing dental restorative materials in increasing the fluoride ion density in enamel and dentin adjacent to the restoration. This fluoride release property may also confer some cariostatic effects on the materials, since fluoride uptake leads to increased resistance of interfacial enamel and dentin to acid. Indeed, various in vitro studies have shown that improved fluoride-containing materials exhibited an inhibitory effect on the development of marginal demineralization following acid exposure. The use of fluoride-releasing materials is thus advocated for the prevention of initial or secondary caries around restorations.

Of late, various fluoride-releasing adhesive systems and composite resins have been developed. In particular, more recently developed fluoride-releasing one-step adhesives contain specific fluoride sources that impart enhanced fluoride-releasing ability to the adhesives. For example, AB contains PEM-F (Penta Methacryloxyethyl cycophosphazene Mono Fluoride) and SI contains S-PRG filler. We have already reported that S-PRG- and PEM-F-containing materials released greater amounts of fluoride than conventional materials which contained fluoridated polymers. S-PRG-containing adhesives released fluoride from S-PRG filler, and recharged fluoride from fluoride mouth rinse. As for AB—the PEM-F-containing adhesive material, it did not show any degradation while releasing fluoride at a steady rate.

Findings of the present study clearly showed that SI released fluoride over a considerably long time period of time (Fig. 1). Moreover, SI specimens clearly showed fluoride uptake by enamel and dentin, which was particularly evident in the dentin specimens following 60 days of water immersion (Figs. 2 and 3). Fluoride ion density in enamel and dentin were different between AB and SI. In particular, fluoride uptake by dentin in SI specimen was clearly observed after the specimen was stored in distilled water for 60 days (Fig. 3). The fluoride uptake layer was more clearly seen in dentin than in enamel, especially with prolonged immersion time. This finding correlated with our previous findings on other types of fluoride-containing materials, and it could be attributed to the structural and compositional differences between enamel and dentin, namely: (1) tubular structure of dentin differs from that of enamel; (2) size and density of apatite crystals in enamel are greater than those in dentin; and 3) water content in dentin is much higher than that of enamel.

Although AB released a considerable amount of fluoride (Fig. 1), a very low level of fluoride uptake by enamel and dentin was observed in all AB specimens. This could be due to a character of PEM-F, such that fluoride uptake by enamel and dentin is not as easy. In the same vein, difference in fluoride uptake between SI and AB specimens could partially be explained by the difference in fluoride releasing component, thereby suggesting that S-PRG filler might have some advantageous influence on fluoride uptake in the tooth structure.

Moreover, acid resistance zones were clearly formed adjacent to the cavity margins of AB and SI specimens (Fig. 4). Indeed, the formation of acid resistance zones further illustrated the important influence of the fluoride-releasing property of these adhesive materials. Hence, in terms of improving acid resistance in tooth structures, the effectiveness of fluoride-containing adhesive materials (SI) was clearly far more superior to that of adhesive materials which did not contain fluoride (GB).

In this study, it was found that apart from the advantage of easy application of one-step adhesives,
the fluoride content was also effective in improving the resistance of enamel and dentin to acid. With these newly developed, fluoride-containing one-step adhesives, it is indeed most beneficial that they are able to prevent secondary caries while at the same time perform their dental restoration role.

CONCLUSIONS

This in vitro study demonstrated that a newly developed S-PRG filler-containing one-step adhesive had fluoride-releasing property that led to uptake of fluoride by enamel and dentin adjacent to the adhesive. Moreover, the corresponding areas showed decreased demineralization following acid exposure. Although these results may not necessarily be extrapolated to the clinical situation, they nonetheless support the notion that fluoride-releasing resin adhesives have some inhibitory effects on the demineralization process of enamel and dentin, and thus contribute to inhibiting recurrent caries formation. This notion may be further established by elucidating the mechanism of action, and in particular, the optimum amount and concentration of fluoride for caries inhibition.

REFERENCES


