Fluoride release and dentin caries inhibition adjacent to resin-modified glass-ionomer luting cements

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The objectives of this investigation were to compare the fluoride release and caries inhibition ability of resin-modified glass ionomer (RMGI) luting cements. Methods: Materials used for dentin caries inhibition were 3M ESPE RelyXTM Luting Plus Cement (RLP) paste-paste RMGI, RelyXTM Luting Cement (RLC) powder-liquid RMGI and FiltekTM Z250 composite/ Adper™ Single Bond adhesive (ZSB). Rectangular slots (6×2×1 mm) were prepared in 24 dentin blocks cut from 8 bovine roots, and filled with ZSB, RLC, and RLP. The specimens were immersed in 10 ml acetic acid solution pH 5.0 (37ºC, 3 wks) to create artificial dentin lesion, then sectioned into 400 µm slices and subjected to microradiography. Mineral loss (∆Z) was calculated from mineral profiles at 0.5 and 1.0 mm from the material margin, and statistically analyzed (ANOVA, Fisher's, p<0.05). Fluoride release from cured cement discs (20×1 mm; n=8) in distilled water was measured after 1, 7, 14, 28, 90, 180, and 365 day using F-specific electrode and TISAB buffer. A conventional GIC, Ketac TM Cem, was used as a control for this experiment. Results: Z250 is not fluoridated and did not exhibit F release. The sustained F release from RLP and RLC were comparable to each other and higher than from the control conventional GIC. Both RMGIs showed pronounced inhibition zones in dentin adjacent to cement margin. ∆Z values at 0.5 mm and 1.0 mm from RLP and RLC were significantly less compared to ZSB. ∆Z values were not significantly different in RLP and RLC groups at 1.0 mm. Conclusion: Both the powder-liquid and paste-paste RMGIs, RelyX Luting and RelyX Luting Plus cements respectively, released comparable amounts of fluoride, and, in contrast to the composite, demonstrated inhibition zones at the adjacent dentin when subjected to in vitro demineralization. Both cements inhibited mineral loss in dentin compared to the composite up to 1.0 mm adjacent to the bonded interface. (Chin Dent J, 24(3): 127-133, 2005)

Key words: caries inhibition, dentin demineralization, fluoride release, luting cement, resin-modified glass-ionomer.

Restoration failure and replacement is an unresolved problem in dental practice1. The main reason for such failure is secondary caries, which has been reported to be responsible for 40 to 60% of restoration replacements1-4. Although risk factors for primary and secondary caries are well known theoretically, patient compliance and host factors can hinder the accurate prediction of restoration failure. Fluoride-releasing materials are indispensable tool to lower the risk of caries development5. Among the vast array of restorative materials available at present, glass ionomer cements (GICs) are the greatest source of fluoride available to the surrounding tooth structures5,6. Studies have shown that glass ionomers inhibit demineralization of the surrounding tooth structures in vitro5,10, in situ11,12 and provide protection against recurrent caries under clinical condition for patients with high caries risk13-16.
This can be attributed to the ability of glass ionomer cements to inhibit demineralization and enhance remineralization through release of fluoride to the adjacent tissue and surrounding fluid.

Resin-modified glass ionomer (RMGI), were developed to provide the advantages of preferred handling and setting characteristics, while maintaining the sustained fluoride release behavior and good clinical adhesion of conventional GICs. Standard RMGIs are usually dispensed in a powder-liquid form. To further improve the handling characteristics, paste-paste delivery system has been introduced. One recent paste-paste RMGI system is the RelyX Luting Plus system (3M ESPE, St Paul, MN) delivered through a convenient metered double-barreled dispenser called the “clicker”. The unique “clicker” delivery not only provides easy dispensing and easy mixing, but the consistent dosing should facilitate the material to achieve its optimal properties.

One of the applications of GICs is as a luting cement in fixed prosthodontics. Apart from the capability of releasing fluoride to protect against secondary caries, RMGI luting cements have advantage over other types of cement in that it can adhere to tooth structures without the need for separate etching or conditioning steps. The self-adhesive property leads to minimal microleakage, and thus diminishes post-operative sensitivity. The manipulative procedure is fast and simple, also due to less concern for isolation, moisture control and anesthesia. In addition, GI luting cements exhibit good mechanical properties, low setting stress, and have evidence to strengthen core ceramics.

While there have been several reports on the caries inhibitory property of GIC and RMGI restoratives there are hardly any published studies on similar efficacy of luting cements. The first objective of this study was to compare the fluoride release of the paste-paste RMGI luting cement with powder-liquid RMGI and conventional GIC. The second objective was to compare in vitro caries inhibition ability of these RMGI luting cements to a composite/adhesive system.

**MATERIALS AND METHODS**

**Fluoride release**

Materials used in the fluoride release experiment are shown in Table 1. Three disk-shape specimens (20 mm in diameter and 1.0 mm in thickness) were prepared from each material using a Plexiglas split mold. The specimens were allowed to mature in a 95%-relative-humidity chamber for 1 h at 37°C before being removed from the mold and weighed on an analytical balance. Each specimen was immersed separately in 25 ml deionized water at 37°C. A fluoride ion-selective electrode (Orion model 96-09-00, Orion) was used to quantify the amount of fluoride released from each specimen into the deionized water. The electrode was calibrated with fluoride standards and the ionic strength was controlled by TISAB buffer. Fluoride released, calculated as µg F/g of specimen, was measured after 1, 7, 14, 28, 90, 180, and 365 days as shown in Figure 1.

<table>
<thead>
<tr>
<th>Material</th>
<th>Product name</th>
<th>Manufacturer</th>
<th>Lot number</th>
<th>Composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conventional powder-liquid GI</td>
<td>Ketac Cem (KC)</td>
<td>3M ESPE</td>
<td>138393</td>
<td>Powder: FAS glass Liquid: polyalkenoic acid, water, tartaric acid</td>
</tr>
<tr>
<td>Powder-liquid RMGI luting cement</td>
<td>RelyX™ Luting Cement (RLC)</td>
<td>3M ESPE</td>
<td>20021227</td>
<td>Powder: FAS glass, opacifier, redox catalysts Liquid: Methacrylated polyalkenoic acid, HEMA, water, tartaric acid, stabilizers</td>
</tr>
<tr>
<td>Paste-paste RMGI luting cement</td>
<td>RelyX™ Luting Plus Cement (RLP)</td>
<td>3M ESPE</td>
<td>A6805 B6802</td>
<td>Paste A: FAS glass, water, HEMA, redox catalyst, opacifier Paste B: Methacrylated polyalkenoic acid, water, HEMA,</td>
</tr>
<tr>
<td>Restorative composite</td>
<td>Z250</td>
<td>3M ESPE</td>
<td>3WR</td>
<td>Zr/Si Filler, BisGMA, TEGDMA, BisEMA6, UDMA, photoinitiator system</td>
</tr>
<tr>
<td>Dentin adhesive</td>
<td>Adper™ Single Bond</td>
<td>3M ESPE</td>
<td>7AT</td>
<td>Etching gel: phosphoric acid, water, fumed silica, colorant Adhesive: Methacrylated polyalkenoic acid, water, ethanol, HEMA, BisGMA, UDMA, GDMA, photoinitiator system</td>
</tr>
</tbody>
</table>
Dentin caries inhibition of glass-ionomer

24 dentin blocks were cut from eight roots of extracted bovine incisors (Figure 2A). Rectangle slots (6×2×1 mm) were prepared in each dentin specimen using #56 carbide bur in high-speed hand piece with copious amount of water. The slots (n=8) were filled with Filtek™ Z250 composite with Adper™ Single Bond adhesive (ZSB), RelyX™ Luting Cement (RLC) powder-liquid RMGI, or RelyX™ Luting Plus Cement (RLP) paste-paste RMGI, and finished with Soflex™ discs (3M ESPE, St Paul, MN). Material information is shown in Table 1.

The specimens were coated all around with acid-resistant nail varnish except the material and adjacent dentin as shown in Figure 2B. Artificial caries lesions were created by immersing the specimen in 10 ml of demineralization solution at 37°C. The demineralizing solution, modified from Mukai et. al.18, contained 50 mM acetic acid, 1.5 mM CaCl₂, 0.9 mM KH₂PO₄, 0.1 ppm F. The pH was adjusted to 5.0 with 1 M KOH. After three weeks, the specimens were removed from the demineralizing solution, rinsed with distilled water, embedded in acrylic resin and cut into thin slices (Figure 2C) using a diamond blade (Isomet™ Low Speed Saw, Buehler, Lake Bluff, IL) with water coolant. Each specimen yielded 3 to 6 dentin slices with approximate thickness of 500 µm. The dentin slices were ground by hand on wet 600 grit SiC paper (Buehler) to achieve a thickness of approximately 400 µm. Figure 2D is the diagram of the specimen ready for microradiography, showing orientation of the restored cavity and the demineralized lesion on adjacent dentin.

The specimens along with an aluminum step wedge were arranged between moist filter paper (Whatman, W&R Balston Limited, UK) to prevent dehydration. Microradiograph was obtained with a Picker ‘Hotshot’ X-ray machine (Picker Industrial Products, Cleveland, OH) operating at 12 kV and 1 mA for the exposure time of 54 s, using Ultra-speed Dental Films (Kodak DF-58, Eastman Kodak Company, Rochester, NY).

The image of the microradiograph was captured...
At 100x magnification under a stereomicroscope (Olympus SZH10, Japan) with CCD camera (Sony DXC-151A, Japan). The light intensity was standardized by adjusting the light source so that an unexposed X-ray film reached a particular luminance value, which allowed a constant light incident. Microdensitometry was carried out using an image analysis software Optimas™ 5.2 (Optimas™ Corporation, Edmonds, WA).

The line luminance function in the software provides a gray scale profile across the area of 0.25 mm width. Each line was scanned from lesion surface to underlying sound dentin (0.5 mm length). Two measurements were done at 0.5 and 1 mm adjacent to the material. Mineral content (volume % mineral) was calculated from the gray value (radiopacity) of the microradiograph calibrated with the aluminum step wedge\(^{19-21}\). A mineral profile was constructed from the mineral content as a function of distance from the lesion surface to the underlying sound dentin. Amount of mineral loss or \(\Delta Z\) (volume % mineral-µm) is the integration of the area between the mineral profile of the lesion and the average volume % mineral that is extrapolated from the underlying sound dentin. Custom software was written to calculate \(\Delta Z\) from each mineral profile by defining the underlying sound dentin to contain 45 volume % mineral as an internal standard.

\(\Delta Zs\) at 0.5 and 1 mm from the material margin were compared between the three materials using a one-way ANOVA followed by Fisher’s test at a significant level of 0.05.

**RESULTS**

**Fluoride release**

The cumulative amount of fluoride release from Z250, the two RMGIs, and the conventional GIC control KC at 1, 7, 14, 28, 90, 180, and 365 days was plotted vs time in Figure 1 and presented in Table 2. Figure 1 shows the amount of cumulative fluoride release from various glass ionomer cements plotted against observation periods up to 1 year. Z250 is not fluoridated and did not liberate fluoride. Both the new paste-paste RMGI, RLP, and the earlier version powder-liquid RMGI, RLC, released comparable amounts of fluoride that are considerably higher than from the conventional GIC Ketac Cem.

**Dentin demineralization**

The subsurface lesion developed on dentin surface next to the material is seen as a dark (radiolucent) area covered by a radiopaque layer. Figure 3 depicts microradiographic images and corresponding mineral profiles of three specimens cut from the same tooth. Dentin lesion adjacent to restorative composite (ZSB) was extensive with very thin surface layer. A wedge-shape area is observed in close proximity to the composite in some cases. In the two glass ionomer groups (RLC and RLP), the lesions were not as deep as in the composite group. The surface layer of the lesions adjacent to RLC and RLP was thick and well-defined. In addition, pronounced inhibition zones where dentin caries could not develop was detected in close proximity to

<table>
<thead>
<tr>
<th>Days</th>
<th>Material</th>
<th>Z250</th>
<th>RLC</th>
<th>RLP</th>
<th>KC</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Z250</td>
<td>0</td>
<td>882 ± 29</td>
<td>809 ± 7</td>
<td>361 ± 27</td>
</tr>
<tr>
<td>7</td>
<td>RLC</td>
<td>0</td>
<td>1970 ± 34</td>
<td>1740 ± 84</td>
<td>849 ± 73</td>
</tr>
<tr>
<td>14</td>
<td>RLC</td>
<td>0</td>
<td>2747 ± 90</td>
<td>2325 ± 35</td>
<td>1276 ± 91</td>
</tr>
<tr>
<td>28</td>
<td>RLC</td>
<td>0</td>
<td>3826 ± 94</td>
<td>3037 ± 64</td>
<td>1889 ± 120</td>
</tr>
<tr>
<td>90</td>
<td>RLC</td>
<td>0</td>
<td>5792 ± 324</td>
<td>5089 ± 339</td>
<td>2967 ± 167</td>
</tr>
<tr>
<td>180</td>
<td>RLC</td>
<td>0</td>
<td>7693 ± 442</td>
<td>6987 ± 445</td>
<td>3870 ± 240</td>
</tr>
<tr>
<td>365</td>
<td>RLC</td>
<td>0</td>
<td>9603 ± 650</td>
<td>8890 ± 682</td>
<td>4462 ± 350</td>
</tr>
</tbody>
</table>

Table 2. Cumulative fluoride release in µg F/g material (mean ± SD) from restorative composite Z250, RelyX™ Luting Cement (RLC), and RelyX™ Luting Plus Cement (RLP) and Ketac Cem (KC) after 1, 7, 14, 28, 90, 180, and 365 days.
In the corresponding mineral profiles at 0.5 and 1.0 mm from the cavity margin, the faint radiopaque surface layer in ZSB group is seen as a small peak followed by the low mineral content body of the lesion, then the profile gradually rises to approach sound dentin. The well-developed surface layer in the RMGI groups is evidence by a large peak in the mineral profiles. The mineral profiles also show that lesions at 1.0 mm are deeper than lesions at 0.5 mm.

The amount of mineral loss (ΔZ) at 0.5 and 1.0 mm from the material margin is shown in Table 3. Significant differences in mineral loss were found among all three materials at 0.5 mm distance (ANOVA and Fisher’s test, p < 0.05). At 1 mm from the cement margin, the differences between RLP and RLC were not significant (ANOVA and Fisher’s test p > 0.05).

In other words, both glass ionomer cements showed higher caries inhibitory effect than composite at 1 mm periphery of the material.

**DISCUSSION**

In the present study, microradiography has been modified from a standard methodology by using a periapical X-ray film instead of a holographic film. The relatively coarse grain of the periapical film was compensated by averaging the radiopacity across an area of 0.25 mm, i.e., the mineral profile at 0.5 mm represents the measurement from 0.375 to 0.625 mm. Low energy x-ray (12 kV) was used in order to permit high absorption by the specimen, thus regions with slight differences in mineral content would be distinguishable. The average mineral content of

**Table 3. The amount of mineral loss (ΔZ; Mean ± SD) at 0.5 and 1.0 mm from the margin of materials, Filtek™ Z250/Adper™ Single Bond adhesive (ZSB), RelyX™ Luting Cement (RLC), and RelyX™ Luting Plus Cement (RLP)**

<table>
<thead>
<tr>
<th>Materials</th>
<th>ΔZ (VPM-µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.5 mm</td>
</tr>
<tr>
<td>ZSB</td>
<td>7641 ± 621</td>
</tr>
<tr>
<td>RLC</td>
<td>4267 ± 1322</td>
</tr>
<tr>
<td>RLP</td>
<td>5435 ± 430</td>
</tr>
</tbody>
</table>

Same letter denotes mean values that are not significantly different for each distance (ANOVA, Fisher’s, p>0.05).
sound dentin computed by this method was 45.52 vol % which is comparable with the established value of 47 %.

Considering high prevalence of restoration failure and replacement with secondary caries as a main cause, restorative material with anticariogenic potential is crucial in dental armamentarium. Glass ionomer cements have shown the anticariogenic properties in several in situ and clinical studies. In a cross-over in situ study, both conventional and resin-modified glass ionomers were able to reduce enamel demineralization under a severe cariogenic challenge where the enamel blocks were dipped in sucrose solution extraorally 8 times a day and the subjects used non-fluoridated dentifrice. Another in situ study also showed less demineralization of enamel adjacent to resin-modified glass ionomer restorative materials than composite group in an interproximal crown model after 4 weeks without fluoride dentifrice.

The anticariogenic effect of glass ionomers is remarkably crucial in high-risk patients. Recurrent caries reductions around cervical restorations for conventional and resin-modified glass ionomers relative to composite were greater than 80% in xerostomic patients with less compliance to fluoride supplementation. Similarly, less caries developed at margins of glass ionomer restorations compared with amalgam after 2 years in xerostomic patients who did not routinely use topical fluoride.

The ability to release fluoride over a prolonged period of time and improve resistance against secondary caries in coronal and root surfaces make glass ionomers a good candidate as luting agent in fixed prosthodontics. In the present study, the new paste-paste RMGI luting cement (RLP) exhibited similar sustained fluoride release as its precursor powder/liquid material (RLC). The total level of fluoride release from both RMGIs was higher than from a classical conventional GIC. Glass ionomer luting cements like the ones present here, which have shown potential to intervene with the development of dentin caries, can be viewed as therapeutic material conforms to the emerging concept of minimally invasive dentistry.

CONCLUSION

In conclusion, RelyX Luting and RelyX Luting Plus cements released comparable amounts of fluoride. The fluoride release was greater than that of conventional GICs studied. When subjected to in vitro demineralization, both luting cements demonstrated inhibition zones adjacent to the cavity margin in contrast to the composite. RelyX and RelyX Plus luting cements showed lower mineral loss in dentin compared to the composite up to 1 mm adjacent to the bonded interface. Clinically these RMGI luting cements may help inhibit the development of caries in the cement proximity. This is especially beneficial for compromised marginal areas that are under cariogenic risks.

REFERENCES


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