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The purpose of this study was to investigate the effects of silanized spherical silica fillers (SF) on the immediate and 24-hour marginal gaps of resin-modified glass-ionomer cement (RMGIC) in tooth cavities. In correlation with marginal gap formation in the tooth cavity, these influencing factors were also examined: marginal gap and setting shrinkage of cement in the Teflon mold, as well as the shear bond strength to tooth substrate. Moreover in correlation with caries prevention, fluoride release was examined too.

In this investigation, the fillers were mixed into the RMGIC powder (Fuji II LC EM). Untreated spherical silica filler (UF)-added RMGIC was used as a comparison. When compared with the control (i.e., original RMGIC), the addition of SF significantly decreased immediate marginal gap in tooth cavities and setting shrinkage in Teflon mold up to 63% and 66% respectively. Fluoride release was significantly reduced too. Apart from these results, this study showed that addition of 5 wt% SF increased the shear bond strength to human enamel and dentin.

Key words: Resin-modified glass-ionomer cement, Silanized spherical silica fillers, Marginal gap

INTRODUCTION
Glass-ionomer cements (GICs) possess several beneficial properties, including physicochemical adhesion to tooth structures and release of fluoride ions. Newer versions of light-cured glass-ionomer, which are known as resin-modified glass-ionomer cements (RMGICs), combine the favorable properties of GICs with those of resin composites. These include the inherent adhesion and cariostatic properties of the former and the command setting behavior, good mechanical properties, and wear resistance of the latter1-4. Compared with conventional analogs, RMGICs have been characterized as having a longer working time, a rapid set, improved appearance and translucency, in addition to a higher early strength along with higher bond strength to enamel and dentin5-9. As a result, RMGICs also have less microleakage than conventional GICs6-8,10,11. However, RMGICs still need surface protection to reduce margin microleakage12, and they also show less fluoride release and caries resistance than GICs13.

In a previous study14, we reported that the addition of spherical silica fillers to a RMGIC powder improved the workability of the cement. The addition of silanized spherical silica fillers (SF) increased the compressive strength, diametral tensile strength and flexural strength, and reduced the water uptake of RMGIC. It was shown through the previous study that SF improved the mechanical properties of the cement more than untreated spherical silica fillers (UF)14. Indeed, the success of SF addition would have had a greater value if it could reduce marginal gap or microleakage and possess other advantages in correlation with the tooth substrate.

Therefore, in this study, the first hypothesis was that the addition of SF to RMGIC would reduce the immediate and 24-hour marginal gaps in tooth cavities. The second hypothesis was that the marginal gap and rate of setting shrinkage in Teflon mold and the shear bond strength of RMGIC would influence marginal gap formation in tooth cavities. The final hypothesis was that the addition of SF would reduce the fluoride release of RMGIC.

MATERIALS AND METHODS

Materials
The RMGIC material used in this study was Fuji II LC EM (powder lot #020521, liquid lot #0204301, GC Corp., Tokyo, Japan) with a recommended powder/liquid ratio (P/L) of 3.0. The detailed composition of the material has not been released. But according to manufacturer’s information, it was given that the powder is fluoro-alumino-silicate glass, and the liquid is composed of methacrylic acid ester, polyacrylic acid, and water.

The silanized spherical silica filler (GC Corp., Tokyo, Japan), which has an average particle diameter of 0.3 μm with γ-methacryloxypropyl trimethoxysilane (γ-MPTS) (KBM 503, Shin-Etsu Chemical Co., Tokyo, Japan), was prepared as previously described15.

The RMGIC powder was modified by initially
mixing it with either the SF or UF at different weight percentages (5%, 10%, and 20%) before mixing it with Fuji II LC EM liquid. The prepared cement powders are described as SF5, SF10, SF20, UF5, UF10, and UF20—hence indicating the type of filler added and filler content in weight percentage. Both the mixing time and preparation time were 30 seconds each. As for the P/L, each one was chosen according to the maximum compressive strength value of each cement in the previous study. Fuji II LC EM was mixed with P/L=3.0 as the control and with P/L=3.6 as the maximum compressive strength of the original RMGIC (FLCEM). SF5, SF10, and SF20 were mixed with P/L of 4.0, 4.4, and 4.0; while UF5, UF10, and UF20 were mixed with P/L of 4.4, 4.4, and 4.0, respectively.

A visible-light curing unit (New Light VL-II, GC Corp., Tokyo, Japan, irradiated diameter: 10 mm) was used for activating the specimens, and close contact was ensured between exit window of the lamp and matrix. The light intensity was checked and maintained at 450 mW/cm² using a radiometer (Demetron/Kerr, Danbury, CT, USA).

Although much information has been generated on the use of bovine teeth, the use of human teeth is a preferred option. A total of 320 human premolars, extracted for orthodontic reasons, were used to measure the marginal gap in tooth cavities and the shear bond strength to tooth tissues in this study. After extraction, the teeth were stored immediately in distilled water at about 4°C within 3 months before use. Since occlusal dentin tends to give lower bond strengths than proximal or buccal dentin, and that dentin tubule orientation and location significantly influence the results of mechanical strength tests, proximal flat enamel or dentin surfaces were used in this study.

All procedures, except for cavity preparation and mechanical testing, were performed in a thermo-hygrostatic room maintained at 23±0.5°C and 50±2% relative humidity. The results were analyzed statistically using ANOVA, the t-test, and Mann-Whitney U-test.

**Marginal gap in tooth cavities**

Human premolars (N=10 for each material and condition) were embedded in a slow-setting epoxy resin (Epofix Resin, Struers, Copenhagen, Denmark). Flat enamel surfaces on the proximal area of the teeth were obtained by grinding with wet silicon carbide paper (800). The cylindrical cavity on the coronal region of each tooth was prepared to a depth of approximately 1.5 mm with a diameter of 3.5 mm with a tungsten carbide bur (200,000 rpm) and a fissure bur (8,000 rpm) using water spray. The dimensions of the cavity were measured using a vernier caliper (U39818, Mitutoyo, Kawasaki, Japan). Each cavity was treated with a Cavity Conditioner (Lot #0205J01, GC Corp., Tokyo, Japan) for 10 seconds according to manufacturer's instructions, rinsed thoroughly with distilled water, air-sprayed, and filled with the material using a syringe tip (Centrix C-R Syringe System; Centrix, Shelton, CT, USA). Covered with a plastic strip, the material was light-cured for 20 seconds. Surfaces were polished immediately after light activation or after storage in distilled water at 37°C for 24 hours. Excess filling material was removed by wet grinding with a wet silicon carbide paper (#1000), followed by polishing using an aqueous slurry of 0.3 μm aluminum oxide (Alfa Micropolish, Buehler Ltd., Lake Bluff, IL, USA) and thorough rinsing with distilled water. Each restoration margin was inspected under a traveling microscope (400×, Measeroscope, MM-11, Nikon, Tokyo, Japan). The maximum gap width and opposing width (if any) between the cement and cavity wall were measured, and the sum of these two values was expressed as the marginal gap in that tooth cavity. As some specimens had zero marginal gaps, especially after 24-hour water storage, the overall sum of 10 specimens examined per material was calculated as the total marginal gap for the group.

**Setting shrinkage in Teflon mold**

Teflon does not react with the filled material. Therefore Teflon molds of the same diameter and depth were prepared to measure the degrees of setting shrinkage (immediately after set) and hygroscopic expansion (after 24-hour water storage), as well as to compare the marginal gap width in the tooth cavity. The Teflon mold was placed on a silicon oil-coated glass plate and a matrix strip that would not react with or bind to the filling material. Each mold was filled with the material using a syringe tip, then covered with a plastic strip and light-cured for 20 seconds. Immediately after setting or after 24-hour storage in distilled water at 37°C, any excess material was removed. The maximum gap width and opposing width between the cement and the Teflon cavity were measured, and the sum of these two values was expressed as the marginal gap in the Teflon cavity. The degree of setting shrinkage was determined using the formula below:

\[ S = \left( \frac{G}{d} \right) \times 100\% \]

where S was the setting shrinkage, G the marginal gap, and d the diameter of the Teflon mold. Ten specimens were prepared for each material and their conditions investigated.

**Shear bond strength to enamel and dentin**

The shear bond strength to enamel and dentin, which comprise the cavity wall, was measured to evaluate the bonding effect between the filling material and the cavity. Bond strength to flat enamel and dentin
surfaces was determined both immediately after light activation and after 24-hour distilled water storage at 37°C. Specimens (N=10 for each material and condition) were obtained from human premolars embedded in the slow-setting epoxy resin. Proximal flat enamel or dentin surfaces were obtained by grinding with a wet silicon carbide paper (#1000) followed by treatment with the Cavity Conditioner for 10 seconds. To prepare the shear bond strength samples, a cylindrical Teflon split mold (3.6-mm diameter × 2.0-mm height) was used to minimize the stress exerted on the specimens during their retrieval. Each material was placed into the Teflon mold set on the enamel or dentinal surface using a syringe tip, then hardened by light-curing. The specimens thus obtained were mounted on a universal testing machine (Autograph DCS-2000, Shimadzu, Kyoto, Japan) and shear stress was applied at 0.5 mm/min, with a maximum external force of 50 kgf (490 N). After shear bond strength measurements were taken, the fractured surfaces were analyzed using a light microscope (4×, SMZ-10, Nikon, Tokyo, Japan) to ascertain the nature of fractures.

Fluoride release measurement
The release of fluoride from SF20, UF20, and FLCEM was measured. All specimens were mixed for 30 seconds using a plastic spatula on a mixing pad, syringe-loaded into a cylindrical Teflon split mold (d=20.0 mm, h=0.5 mm), and covered with a glass plate and clamped. After being light-cured for 20 seconds at five overlapping sites, the samples were removed from the mold, placed in polyethylene vials with 25 ml of distilled water, and stored in an incubator at 37°C for 1 day, 2 days, 3 days, 1 week, and 2 weeks. For each material and each time period, three samples were prepared. After each specified period, the samples were removed from the medium, and 10 ml of the medium was transferred to another vial and mixed with 1 ml of TISAB III solution (Thermo Orion, Beverly, USA). The amount of fluoride ions released from the RMGIC sample in this solution was measured using a pH/ion meter (F23-S937, Horiba, Kyoto, Japan). Then, using a SigmaPlot 8.0 program (SPSS, Chicago, Illinois, USA) by placing the cumulative fluoride release ([F]₀, mg/L) as a function of time (t, day), the data were fitted to a single rectangular II, 3-parameter equation as follows:

\[ [F]₀ = [F]₀/t + (t/t₁/₂ + t) + α t \]

where \([F]₀\) was the total amount of fluoride to be released, \(t₁/₂\) the time needed to release half of this total amount, and \(α\) the kinetic parameter.

RESULTS
Marginal gap in tooth cavities
Table 1 lists the effects of spherical silica fillers on the marginal gap in tooth cavities. The marginal gaps of all SF-added RMGICs were 67% of the control's or less. The marginal gap of SF10 was 63% of the control's or 83% of FLCEM's, while that of UF10 was 62% of the control's or 81% of FLCEM's. Comparing the sums of the immediate gap width using a non-parametric t-test, all materials with fillers showed significant differences from the control and FLCEM (p<0.05). However, there were no significant differences among SF5, SF10, and UF10; or among SF5, SF10, and UF20; or between SF20 and UF5. After 24-hour storage in distilled water, there were no significant differences among the control, FLCEM, and UF20, as well as

<table>
<thead>
<tr>
<th>Material</th>
<th>P/L</th>
<th>The sum of marginal gaps for all 10 specimens ((µm))¹</th>
<th>α value²</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Immediately</td>
<td>After 24 hours</td>
</tr>
<tr>
<td>SF5</td>
<td>4.0</td>
<td>74 [0] (4–10)¹b</td>
<td>6 [4] (0–1)¹e</td>
</tr>
<tr>
<td>SF10</td>
<td>4.4</td>
<td>73 [0] (5–11)¹b</td>
<td>6 [4] (0–1)¹e</td>
</tr>
<tr>
<td>SF20</td>
<td>4.0</td>
<td>77 [0] (2–11)¹c</td>
<td>6 [4] (0–1)¹e</td>
</tr>
<tr>
<td>UF5</td>
<td>4.4</td>
<td>82 [0] (5–13)¹c</td>
<td>6 [4] (0–1)¹e</td>
</tr>
<tr>
<td>UF10</td>
<td>4.4</td>
<td>71 [0] (2–11)¹a+</td>
<td>6 [4] (0–1)¹e</td>
</tr>
<tr>
<td>UF20</td>
<td>4.0</td>
<td>77 [0] (5–12)¹c</td>
<td>7 [3] (0–1)¹h</td>
</tr>
<tr>
<td>FLCEM</td>
<td>3.6</td>
<td>88 [0] (4–14)¹d</td>
<td>7 [3] (0–1)¹h</td>
</tr>
<tr>
<td>Control</td>
<td>3.0</td>
<td>115 [0] (7–17)¹f</td>
<td>7 [3] (0–1)¹h</td>
</tr>
</tbody>
</table>

SF=Silanized spherical silica filler; UF=Untreated spherical silica filler
FLCEM=Fuji II LC EM mixed with P/L 3.6; Control=Fuji II LC EM mixed with P/L 3.0, n=10
¹ = Number of specimens with no gaps
² = Range of values
1: Identical letters indicate no significant differences among the materials analyzed using the t-test (p>0.05, non parametric)²³
2: Significant difference, analyzed using the Mann-Whitney U-test between two sums

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among all other spherical filler-added RMGICs. The Mann-Whitney U-test showed a significant difference (α < 0.05) between the immediate and 24-hour values for each material. Compared with the control, the immediate gap width mean values of all spherical filler-added RMGICs showed a significant difference (Fig. 1).

Setting shrinkage in Teflon mold
Table 2 lists the effects of spherical silica fillers on the setting shrinkage of RMGIC in the Teflon mold. At the immediate time point, there were no significant differences among FLCEM, UF5, UF20, and SF20. All materials, except FLCEM, showed a significant difference (p<0.05) in comparison with the control. The immediate setting shrinkage of all SF-added RMGICs was 73% of the control's or less. SF10 had a setting shrinkage of 0.62±0.08%, of which its value was 66% of the control's or 78% of FLCEM's. However, there were no significant differences among the 24-hour setting shrinkage values in the Teflon mold. There was a significant difference (p<0.05) though, between the immediate and 24-hour values for each material. Compared with the control, the immediate gap width mean values of all spherical filler-added RMGICs showed a significant difference (Fig. 1). There was also a significant difference between the immediate marginal gaps in the tooth cavity and in the Teflon mold (p<0.05).

Shear bond strength to enamel and dentin
Table 3 lists the effects of spherical silica fillers on the shear bond strength of RMGIC to enamel. Examining the immediate shear bond strengths to enamel, there was a significant difference between SF5 with the control and with UF20 (p<0.05). After 24-hour storage, there was a significant increase (p<0.05) in shear bond strength to enamel, and SF5 showed the highest shear bond strength value. Table 4 lists the effects of spherical silica fillers on the shear bond strength of RMGIC to dentin. There were no significant differences among the immediate shear bond strengths to dentin. However, after 24-hour storage, SF5 and FLCEM showed a significant difference in comparison with SF10, SF20, UF10, and UF20. There was also a significant increase (p<0.05) in shear bond strength to dentin after 24-hour water storage. However, the increase was not significant in SF20 and UF20.

During the course of shear bond strength tests, the majority of failures occurred in RMGIC itself rather than at the RMGIC/tooth interface, which is defined as a cohesive failure. 30,31

Fig. 1 Comparison of immediate marginal gap mean values of RMGICs in the Teflon mold and in the tooth cavity, n=10.
Identical letters indicate no significant difference among the materials analyzed using the t-test for differences among several means (p>0.05).

Table 2 Setting shrinkage in Teflon mold

<table>
<thead>
<tr>
<th>Material</th>
<th>P/L</th>
<th>Immediately (%)</th>
<th>After 24 hours (%)</th>
<th>α value</th>
</tr>
</thead>
<tbody>
<tr>
<td>SF5</td>
<td>4.0</td>
<td>0.64±0.10&lt;sup&gt;a&lt;/sup&gt;</td>
<td>0.15±0.04&lt;sup&gt;d&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>SF10</td>
<td>4.4</td>
<td>0.62±0.08&lt;sup&gt;a&lt;/sup&gt;</td>
<td>0.14±0.04&lt;sup&gt;d&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>SF20</td>
<td>4.0</td>
<td>0.69±0.11&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.16±0.04&lt;sup&gt;d&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>UF5</td>
<td>4.4</td>
<td>0.65±0.09&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.14±0.05&lt;sup&gt;d&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>UF10</td>
<td>4.4</td>
<td>0.63±0.07&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.15±0.06&lt;sup&gt;d&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>UF20</td>
<td>4.0</td>
<td>0.71±0.08&lt;sup&gt;c&lt;/sup&gt;</td>
<td>0.15±0.05&lt;sup&gt;c&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>FLCEM</td>
<td>3.6</td>
<td>0.80±0.08&lt;sup&gt;c&lt;/sup&gt;</td>
<td>0.16±0.03&lt;sup&gt;c&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>Control</td>
<td>3.0</td>
<td>0.94±0.10&lt;sup&gt;c&lt;/sup&gt;</td>
<td>0.17±0.05&lt;sup&gt;c&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
</tbody>
</table>

SF=Silanized spherical silica filler; UF=Untreated spherical silica filler
FLCEM=Fuji II LC EM mixed with P/L 3:6; Control=Fuji II LC EM mixed with P/L 3.0, n=10
Identical letters indicate no significant differences among the materials analyzed using the t-test for differences among several means (p>0.05).<sup>30</sup>
1: Percentage to the diameter of the Teflon mold (inner diameter: 3.5 mm)
2: Significant difference, analyzed using the t-test between two values
Table 3 Shear bond strength of RMGIC to enamel surface

<table>
<thead>
<tr>
<th>Material</th>
<th>P/L</th>
<th>Immediately (MPa)</th>
<th>After 24 hours (MPa)</th>
<th>α value&lt;sup&gt;1&lt;/sup&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>SF5</td>
<td>4.0</td>
<td>7.85±1.57 (8)&lt;sup&gt;a&lt;/sup&gt;</td>
<td>27.47±5.12 (10)&lt;sup&gt;f&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>SF10</td>
<td>4.4</td>
<td>7.34±2.02 (10)&lt;sup&gt;a,b,c&lt;/sup&gt;</td>
<td>22.47±5.31 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>SF20</td>
<td>4.0</td>
<td>6.94±1.43 (10)&lt;sup&gt;a,b,c&lt;/sup&gt;</td>
<td>19.74±4.94 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>UF5</td>
<td>4.4</td>
<td>7.03±1.39 (9)&lt;sup&gt;a,b,c&lt;/sup&gt;</td>
<td>21.17±4.69 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>UF10</td>
<td>4.4</td>
<td>6.42±2.15 (9)&lt;sup&gt;a,b,c&lt;/sup&gt;</td>
<td>18.01±4.86 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>UF20</td>
<td>4.0</td>
<td>6.02±1.02 (6)&lt;sup&gt;e&lt;/sup&gt;</td>
<td>12.13±3.86 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>FLCEM</td>
<td>3.6</td>
<td>7.61±1.65 (9)&lt;sup&gt;a&lt;/sup&gt;</td>
<td>24.78±3.75 (10)&lt;sup&gt;e&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>Control</td>
<td>3.0</td>
<td>6.35±1.72 (8)&lt;sup&gt;b,c&lt;/sup&gt;</td>
<td>23.93±3.41 (10)&lt;sup&gt;e&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
</tbody>
</table>

SF = Silanized spherical silica filler; UF = Untreated spherical silica filler; FLCEM = Fuji II LC EM mixed with P/L 3.6; Control = Fuji II LC EM mixed with P/L 3.0, n=10

Identical letters indicate no significant differences among the materials analyzed using the t-test for differences among several means (p>0.05)<sup>38</sup>

( ): Cohesive failure, among 10 specimens

1: Significant difference, analyzed using the t-test between two values

Table 4 Shear bond strength of RMGIC to dentin surface

<table>
<thead>
<tr>
<th>Material</th>
<th>P/L</th>
<th>Immediately (MPa)</th>
<th>After 24 hours (MPa)</th>
<th>α value&lt;sup&gt;1&lt;/sup&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>SF5</td>
<td>4.0</td>
<td>7.60±2.16 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>13.54±3.60 (10)&lt;sup&gt;d&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>SF10</td>
<td>4.4</td>
<td>7.42±1.49 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>10.28±3.60 (10)&lt;sup&gt;d&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>SF20</td>
<td>4.0</td>
<td>7.88±3.68 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>10.29±3.64 (10)&lt;sup&gt;d&lt;/sup&gt;</td>
<td>NS</td>
</tr>
<tr>
<td>UF5</td>
<td>4.4</td>
<td>7.67±1.57 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>11.54±3.32 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>UF10</td>
<td>4.4</td>
<td>7.45±2.62 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>10.03±2.54 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>UF20</td>
<td>4.0</td>
<td>7.30±2.49 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>8.16±2.44 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>NS</td>
</tr>
<tr>
<td>FLCEM</td>
<td>3.6</td>
<td>6.47±2.39 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>13.48±3.77 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
<tr>
<td>Control</td>
<td>3.0</td>
<td>6.44±2.31 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>10.74±3.27 (10)&lt;sup&gt;b&lt;/sup&gt;</td>
<td>p&lt;0.05</td>
</tr>
</tbody>
</table>

SF = Silanized spherical silica filler; UF = Untreated spherical silica filler; FLCEM = Fuji II LC EM mixed with P/L 3.6; Control = Fuji II LC EM mixed with P/L 3.0, n=10

Identical letters indicate no significant differences among the materials analyzed using the t-test for differences among several means (p>0.05)<sup>38</sup>

( ): Cohesive failure, among 10 specimens

1: Significant difference, analyzed using the t-test between two values

Fluoride release

Fig. 2 shows the effects of spherical silica fillers on fluoride release from RMGIC. SF or UF reduced the release of fluoride. During two weeks’ immersion in distilled water, the fluoride release of SF20 (which fulfilled the equation F=4.80t/(0.01+t)+0.79t) was 49.4% - 57.7% of FLCEM’s (F=12.79t/(0.46+t)+1.10t). For the same period, the value for UF20 (F=15.71t/(1.12+t)+0.68t) was 84.6% - 99.8% of FLCEM’s. In two weeks, the cumulative fluoride release value of SF20 was 15.88±1.89 mg/L, while those of FLCEM and UF20 were 27.87±0.64 mg/L and 23.83±0.64 mg/L respectively.

Analyses using ANOVA revealed that both the addition of spherical silica fillers and the period of immersion in distilled water significantly (p<0.001) influenced fluoride release from RMGIC.
DISCUSSION

Many factors challenge the marginal integrity of GIC restorations. These factors include cavity preparation method\(^{27}\), cavity pretreatment, curing shrinkage\(^{28-30}\), and hygroscopic expansion\(^{22,23,26}\) of the material. For RMGICs, stress development due to cement setting starts immediately after light irradiation is commenced\(^{31}\), and curing shrinkage can create marginal gaps that may contribute to restoration failure\(^{29,30}\). Therefore, water exposure was recommended to control stress development\(^{22,31}\), and it was suggested to delay polishing by at least one day to prevent gap formation at the cement-tooth cavity interface by anticipating the hygroscopic expansion of the restorative material\(^{22,23,31}\). This effect was reported as uptake of water by the RMGIC matrix to form a poly-HEMA complex\(^{30}\).

It was reported that the 24-hour water uptake of SF-added RMGICs was less than that of UF-added RMGICs or the original RMGIC\(^{14}\). However, after 24-hour storage there were no significant differences between the marginal gap widths of SF-added RMGICs and those of UF-added RMGICs, except for UF20. Nonetheless, RMGICs added with either filler type showed significant differences in comparison with the control and FLCEM (Table 1). Although the hygroscopic expansion did not entirely compensate the setting shrinkage in the Teflon mold (Table 2) after 24-hour water storage, the marginal gaps in tooth cavities were remarkably eliminated.

At the immediate time point, the marginal gaps in the Teflon mold were about three-fold of those in the tooth cavity (Fig. 1). Smaller marginal gaps observed in the tooth cavity than in the Teflon mold clearly demonstrated that the cement and cavity walls was an important factor influencing marginal gaps\(^{30}\). For this reason, direct studying of marginal gaps in tooth cavities was a useful simulation of clinical conditions\(^{30}\). However, by examining the setting shrinkage in the Teflon mold, the marginal gap in the tooth cavity could be predicted since there was a linear correlation between the values\(^{30}\) (Fig. 3).

It was reported that resin infiltration into the expanded decalcified layer would prevent microleakage and nanoleakage\(^{24}\). Furthermore, marginal integrity is of primary importance in maintaining the long-term function of resin-based restorations\(^{25}\). At the immediate time point, when compared with the control, the SF- and UF-added RMGICs had significantly less marginal gaps in the tooth cavity and setting shrinkage in the Teflon mold. This occurred due to one or both of the following reasons. First, the SF- or UF-added RMGIC was mixed with higher P/L. The higher the P/L, the smaller the marginal gap\(^{31}\) and the greater mechanical strength of the GIC\(^{14,31,36}\). Second, the spherical form of silica fillers increased the flowability of RMGIC\(^{14}\), lending to better penetration into the decalcified layer and reducing the immediate marginal gap.

Following post-set water uptake, the acidic monomers of RMGIC ionize then react with the glass filler to initiate an acid-base reaction, producing ionic cross-linking\(^{37}\). In these dual setting systems, resin-reinforcement produces higher bond strengths to dental tissues as well as enhanced mechanical strength\(^{26}\). Van Meerbeek et al.\(^{37}\) found a gradual transition of resin concentration in the decalcified superficial dentin. Resin concentration was highest at the top of the hybrid layer, and lowest near the base. If resin did not penetrate through the full depth of the decalcified zone, the non-infiltrated weak collagen layer at the bottom of the zone would perhaps be susceptible to long-term hydrolytic degradation\(^{38}\). Since the majority of shear bond failures were cohesive failures, the interfacial bond formed between the RMGC and the solid substrate was stronger than the cohesive bond within the material itself\(^{37}\). However, at the immediate time point, UF20 showed apparently less cohesive failure than other materials. It was thought to stem from poor bonding of UF to the RMGIC matrix\(^{14}\). As a result, the reaction was delayed, hence affecting the penetration of RMGIC to the decalcified layer. This condition should be studied further.

There was a tendency for the shear bond strength to enamel and dentin to decrease when a large amount of fillers was added. This was clearly shown with the addition of 20 wt% filler, especially in UF50. As mentioned previously, since UF did not bond with the RMGIC matrix\(^{14}\), a large amount of UF may act as an impurity that hinders the reaction in RMGIC. In contrast, SF5 showed higher shear
bond strength to enamel and dentin than those of the control and PLCEM. In addition, up to 10 wt% SF, the RMGICs showed no significant differences in shear bond strength to enamel compared with those of the control and PLCEM. It was thought that an increase in mechanical strength would lead to an increase in bonding ability. Although the 24-hour shear bond strength to dentin did not show a significant difference, supported by the results of mechanical properties in previous study, the SF-added RMGICs showed better mechanical and bond strengths than the UF. These results obviously showed that silanization, which creates siloxane bridges (Si-O-Si) — between the silica surface and the silane molecules — facilitate better interaction between the RMGIC matrix and the filler.

It has been reported that decrease in marginal gap due to increase in P/L would result in increased shear bond strength of the GIC. However, in this study, there was no significant relationship between marginal gap width and shear bond strength, since the filler content of the materials varied. Nonetheless, the shear bond strength results made a great contribution since the SF-added RMGICs were mixed with higher P/L. As mentioned, increasing the P/L will reduce the amount of liquid used and consequently reduce allergies or harmful effects caused by the release of non-polymerized free monomers from the filling materials.

The hydrophobic characteristic of SF may influence both surface diffusion and grain boundary diffusion, and increase the resistance of cement to water uptake. Since the material must first absorb water to initiate any attack on fluoride glass and the dissociation of acid groups, it was not surprising that the addition of SF would reduce fluoride release (Fig. 2). A previous study reported that fluoride has a low rate of diffusion through an organic cross-linked matrix because of strong interaction with aluminum and calcium ions linked to the polyalkenoate chains. The strong interaction between SF and the RMGIC matrix also affected the diffusion rate of fluoride, but fluoride release was not eliminated. This effect should be taken into consideration, because the presence of saliva in vivo may retard the release of fluoride from glass-ionomer filling materials. Although observations were limited to a period of just 14 days in this study, the equation for fluoride release in glass-ionomer cement \[ F_3 = [F]_t / \langle t_{1/2} + t \rangle + a t \], as previously described, accurately approximated the release of fluoride from RMGIC with or without a spherical silica filler \( (r=0.99, p<0.01) \). A similar pattern of release was also obtained, and the release was much greater in the first 24 hours than subsequently. The small but significant initial release was undoubtedly the result of fluoride leaching from glass particles in the surface layer. A previous study showed that \( \gamma \)-MPTS treatment was useful for the regulation of NaF release from bis-GMA/TEGDMA resin. There was evidence that a large amount of fluoride was released from the resin containing NaF powder without \( \gamma \)-MPTS treatment in the initial stage, and the release of fluoride ceased in a relatively shorter time. However, smaller amounts of fluoride were released from the resin containing NaF treated with higher concentrations of \( \gamma \)-MPTS, and the release lasted for longer periods. GICs release and take up fluoride ions. As such, the fluoride release and fluoride recharge capabilities of spherical silica filler-added RMGICs should be examined for a longer period in a further study.

CONCLUSION

The addition of spherical silica fillers to RMGIC significantly decreased the marginal gap and the setting shrinkage in the Teflon cavity. This condition led to a significant decrease in immediate marginal gaps in the tooth cavity. This study also revealed that shear bond strength was not in correlation with marginal gap width and setting shrinkage rate. Nonetheless, if up to 5% of silanized spherical silica fillers were added to RMGIC powder, the shear bond strength of RMGIC to enamel and dentin surfaces would increase. Finally, the addition of spherical silica fillers decreased but did not eliminate fluoride release from RMGIC.

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REFERENCES

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