Chapter 11

Coating glass-ionomer cements with a nanofilled resin

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Abstract

Aim: The objective of this study was to investigate the effect of a nanofilled resin coat on the flexural strength (FS) and the early wear (after 50,000 and 200,000 cycles) of the glass-ionomer cements Fuji IX GP Extra (FIXE) and Ketac Molar Aplicap (KM).

Methods: Specimens were prepared and half of them were coated with G-Coat Plus. The uncoated specimens were used as controls. Flexural strength (n = 10) was evaluated after 24 hours using a three-point-bending test on a universal testing machine (ISO 9917-2). Wear (n = 20) was evaluated after 50,000 and 200,000 cycles using the ACTA wear machine. One-way, two-way ANOVA and Tukey post hoc tests were used to analyze differences in FS and wear.

Results: For FIXE the coat significantly increased the FS and the wear along the two time spans. KM did not show a significant difference in FS with the coat. Improvements in wear were observed only after 50,000 cycles.

Conclusion: Based on these laboratory results, it is concluded that G-Coat Plus is indicated in association with FIXE with the aim to improve the mechanical properties of the former. However, this study is limited to a short-term observation.
Introduction

Glass-ionomer cements (GICs) are widely used in preventive and pediatric dentistry as they release fluoride and have chemical adhesion to dental structures (1). This latter property is believed to be beneficial in terms of resistance to degradation of the interface between GIC and dental substrate (2), which also promotes advantages regarding microleakage and secondary caries formation (2). Moreover, GIC have an expansion coefficient similar to the tooth structure, biocompatibility, chemical setting reaction and fluoride release/uptake, attributing a preventive character to them (1-5). In the late 1990s, the conventional GICs were replaced by the highly viscous GICs, which have a faster setting time and notably higher strengths (6). They are mainly used in single surface restorations and in non-carious cervical lesions. They are also the material of choice for the atraumatic restorative treatment (ART) and for restorations that are at risk of early water contamination (7).

Some GICs characteristics may impose limits to its indication for clinical use. The long setting reaction time and the water sensitivity during setting reaction (first 24 hours) associated with a relatively low fracture strength and high occlusal wear rate in comparison to amalgam and modern resin materials are considered drawbacks of conventional GIC (6, 8-10). The setting reaction of conventional glass-ionomers needs water to form the polyacrylate matrix. This initial stage, which is the clinical setting reaction, occurs within the first 10 min after mixing. The second stage, involving the release of the calcium and aluminium cations within the matrix, is a slower continuation of the acid–base reaction that lasts 24 hours (11). During the first reaction, the material is very sensitive to water uptake, while during the second stage the material is very susceptible to dehydration. This short-term sensitivity to water can result in lower strength of the surface and, as a consequence, in low wear resistance and low early flexural strength. This in turns restricts the full potential of GICs for dental applications (8, 12-14). To avoid problems within the first 24 hours, it is recommended to protect the GIC’s surface. In case the manufacturers do not provide special varnishes, copal cavity varnish, light-cured bonding resins, petroleum jelly, cocoa butter, or even nail varnish can be used as such (11, 15-18). The longer this protective material is in contact with the restoration, the smaller the chance that the material will have its mechanical properties reduced (2). Recently, a new generation of coating for GIC has been introduced. The main difference between this material (G-Coat Plus) and the previous generations of coat for GIC is the content of nanofiller particles. A previous study showed no effect of the G-Coat Plus on the fracture toughness of Fuji IX while fracture toughness of a resin-modified glass-ionomer (Fuji II LC) was positively affected (19). On the other hand, it is expected that the conventional GIC, being more sensitive to water, would be more positively affected by the coat.

In contrast to single-surface, the success rate of multi-surface ART restorations is still not satisfactory (20). Gross marginal defects due to wear and fracture of the restoration are among the most common failure reasons. It is interesting to know if the G-Coat Plus would have a positive influence on the fracture strength and the early wear on the most commonly used highly viscous GIC.

The aim of this study was to investigate the flexural strength and the early wear resistance of highly viscous GICs coated and uncoated with a nanofiller-containing fluid resin. The null-hypotheses tested were that there is no difference in (I) flexural strength and (II) wear between coated and uncoated specimens.
Materials and Methods

The materials used in the study, their manufacturers, batch numbers, shade and expiration dates are shown in Table 1. The GICs were supplied in capsulated form and activated/mixed according to manufacturers’ instructions.

Table 1: List of the materials tested, manufacturers, batch numbers, shade and expiration dates.

<table>
<thead>
<tr>
<th>Material Code</th>
<th>Manufacturer Batch/shade/exp date</th>
</tr>
</thead>
<tbody>
<tr>
<td>GC Fuji IX GP Extra</td>
<td>GC Europe (Leuven, BE) 0609261/A3/2008-09 and 0812151/A2/2010-12</td>
</tr>
<tr>
<td>G-Coat Plus</td>
<td>GC Europe (Leuven, BE) 0610241/-/2008-10 and 0809131/-/2010-09</td>
</tr>
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</table>

Flexural strength

The flexural strength (FS) was determined according to the ISO Standard 9917-2 using 25 x 2 x 2 mm bar-shaped specimens. Twenty specimens were made for each material. The GICs were mixed by a Rotomix (3M ESPE) and set at room temperature for 10 min. After setting, the specimens were removed from the molds and randomly assigned in two groups (n = 10). The control group did not receive any surface protection. The experimental group had one surface coated with G-Coat Plus and light cured for 20 s in three sites along the bar (Elipar Highlight, 3M ESPE, power density of 750 mW/cm²). Both groups were stored in liquid paraffin at 37°C (14). After 24 hours the height and width of the specimens were recorded with a digital caliper. The specimens were subjected to a three-point-bending test on a universal testing machine (Hounsfield Ltd, Redhill, Surrey, UK) at a crosshead speed of 0.75 mm/min. The FS was calculated with the following equation:

\[
FS = \frac{3Fl}{2wh^2}
\]

where \( F \) is the load at fracture, \( l \) the distance between the supports (20.0 mm), \( w \) the specimen width, and \( h \) the specimen height. For the experimental group, the coated surface was placed opposite to the applied load.

Wear

Three-body wear was evaluated with the ACTA wear machine (21). In short, this device consists of two motor-driven cylindrical wheels rolling over each other with a surface slip of 15% inside a bowl containing a third body medium, consisting of a slurry of rice and millet seed shells (pH = 7). Fuji IX GP Extra and Ketac Molar were mixed by a Rotomix (3M
For this experiment, new specimens were prepared to fit the wear machine. A total of 8 specimens were used for this experiment, i.e. two experimental specimens and two control specimens of each material. The specimens were placed into one of these wheels and set at room temperature for 10 min. The specimen wheel consisted of ten compartments, each containing approximately 1 g of cement. After setting, the specimens are removed from the mold and repositioned with cyanoacrylate ester (Super Bonder, Henkel Loctite Products, Rocky Hill, CT, USA). The wheel with the specimens was kept at 37°C and 100% humidity for the whole period of the test. A test run consisted of 200,000 cycles of the specimen wheel (taking approximately 55.5 hours) at a rotational speed of 1 Hz to simulate the chewing frequency.

The surface finishing was done 10 min after mixing by placing the specimen wheel on the fixed axis of the wear machine and a diamond-grinding wheel on the other axis, which was allowed to move slowly into the direction of the specimen wheel. The grinding procedure was carried out under water at 37°C with grinding wheels with grits up to 1000. The grinding wheel was replaced by the 'antagonist wheel', which was pressed against the specimen wheel by a spring force of 15 N and rotated with a surface speed, which differed 15% from that of the specimen wheel (15% slip).

A wear-in run of 10,000 cycles was performed to adapt the two wheel surfaces and the worn-in surfaces were measured by profilometry (A1 - Baseline). A total of four specimens, two from Fuji IX GP Extra (FIXEC) and two from Ketac Molar (KMC), were coated with G-Coat Plus (specimens with C as last letter) and light cured for 20 s 3 times along the specimen (Elipar High-light, 3M ESPE). After coating, the profile was measured again (B1) to establish the profile of the coating. Then, a wear run of 50,000 cycles was initiated. After 50,000 cycles, the profile was re-measured (C1) and followed by another wear run of 150,000 cycles, giving a total of 200,000 cycles. At the end of this run, which was four days after the preparation of the specimens, the final profiles were measured (D1). The procedure is schematically depicted in Figure 1. After completion of the sequence A1 → D1, the whole procedure was repeated (A2, B2, C2, and D2). For this part of the experiment, a fresh coating layer was applied again on the worn surfaces of FIXEC and KMC before the wear runs. Freshly prepared slurries were used for all three sequences.

After each run, ten tracings were taken at fixed positions on the worn surface of each pair of specimen (PRK profilometer No. 720702, Perthen GmbH, Hannover, GE) so the loss of material (µm) could be measured. The positioning of the specimen wheel and the direction in which the surface tracings were taken, are shown in Figure 2.

The wear (in µm) after 50,000 cycles was calculated by subtracting the profile after the wear-in run of 10,000 cycles (A) from the profile after 50,000 cycles (C). The wear (in µm) at 200,000 cycles was calculated by subtracting the profile after the wear-in run of 10,000 cycles (A) and the profile after 200,000 cycles (D). It should be noted that after completion of the first sequence (A1 → D1) a fresh coating layer was applied, before measuring B2, C2, and D2. The coating thickness was calculated by subtracting the profile after the wear-in run of 10,000 cycles (A) from the profile after coating (B).

Scanning electron microscopy (SEM)

Representative cross-section of the specimens from the FS test were selected for SEM images. Replica impressions were taken using President™ JET light-bodied (Coltene-Whaledent, Alsätten, Switzerland) as the impression material. Replicas were made with
Coating glass-ionomer cements with a nanofilled resin epoxy resin (Epo-thin® Buehler Ltd, Lake Bluf, IL, USA) mounted on aluminium stubs, gold-sputtered and examined using a scanning electron microscope (XL 20: Philips, Eindhoven, NL) in order to illustrate the interaction between the GIC and G-Coat Plus.

Figure 1: Top: Schematic representation of the wear after 50,000 cycles (C-A) and after completion of 200,000 cycles (D-A) of the uncoated specimens. Bottom: Schematic representation of coating thickness in μm (B-A), wear after 50,000 cycles (C-A) and after completion of 200,000 cycles (D-A) of the coated specimens after the initial coat.

Statistical Analysis

Two-way ANOVA and Tukey post hoc test were used to test differences in wear of the GICs with or without the coat and the effect of time/experiment. The flexural strength was analyzed with one-way ANOVA and Tukey post hoc test. Pearson correlation test was performed to investigate possible correlation between the materials in both tests. The software used was Sigma Stat 3.1 (SPSS inc., Chicago, USA).

Results

The flexural strength data are summarized in Table 2. One-way ANOVA (F=31.2; p<0.001) and post hoc Tukey’s test (p<0.01) showed a significant improvement on the flexural strength of Fuji IX GP Extra when the G-Coat Plus was applied. For the Ketac Molar no differences were observed between the groups with and without the coat. Pearson correlation test showed no correlation between the materials in both tests.

The SEM micrographs showed that there are micro-mechanical interlocking between both materials and the coat (Figures 3 and 4).
Figure 2: Schematic representation of the various steps in the wear experiment. Upper left: Specimen wheel with specimens cured and glued in the wheel. Middle: Antagonist wheel and specimen wheel rolling over each other in a slurry of rice and millet seed shells to produce 3-body (abrasive) wear. For standard experiments the antagonist wheel is pressed against the specimen wheel by a spring force of 15 N and the surface velocity is adjusted to obtain a slip rate of 15%. The rotational speed of the specimen wheel is fixed at 1 Hz. Upper right: Profile tracings from one unworn reference to the other across the worn surface to measure wear.

Table 2: Mean (SD) flexural strength (F<sub>s</sub>) in MPa for investigated materials. Different letters indicate a statistically significant difference (p<0.01).

<table>
<thead>
<tr>
<th>Material</th>
<th>Flexural strength (F&lt;sub&gt;s&lt;/sub&gt;) without G-Coat Plus</th>
<th>Flexural strength (F&lt;sub&gt;s&lt;/sub&gt;) with G-Coat Plus</th>
</tr>
</thead>
<tbody>
<tr>
<td>FIXE</td>
<td>20.2 (4.1)&lt;sup&gt;c&lt;/sup&gt;</td>
<td>34.9 (6.7)&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>Ketac Molar</td>
<td>44.1 (5.6)&lt;sup&gt;a&lt;/sup&gt;</td>
<td>36.1 (5.8)&lt;sup&gt;ab&lt;/sup&gt;</td>
</tr>
</tbody>
</table>
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Figure 3: SEM micrographs of the specimens’ cross-section of FIXE (A) and KM (B), where it is observed that there are micromechanical interlocking between the coat and the GIC.

Figure 4: SEM micrograph of the specimens’ cross-section of KM. Part of the GIC fell off and the coat was left intact. The arrows indicate areas where the coat penetrated the voids of the GIC.

The wear data are summarized in Table 3 and graphically depicted in Figure 5. The mean coating thickness (SD) for Fuji IX GP Extra was 25.3 (4.7) μm (50,000 cycles) and 48.4 (10.6) μm (200,000 cycles) and for Ketac Molar 26.1 (5.5) μm (50,000 cycles) and 42.4 (9.9) μm (200,000 cycles). Two-way ANOVA showed that time (F=4327.2; p<0.001) and the different GICs with or without coat (F=319.4; p<0.001) had a significant effect on wear. Tukey’s post hoc test (p<0.05) showed that wear decreased significantly after 200,000 cycles comparing with 50,000 cycles for all groups. The coated specimens showed significantly lower wear than the uncoated specimens, except for the Ketac Molar groups after 200,000 cycles, which showed the same wear irrespective of the coating application.
Table 3: Wear in µm after 50,000 cycles (C-A) and after completion of 200,000 cycles (D-A) at two time span. Negative values indicate that there is still presence of coating. Standard deviations in parentheses are from ten tracings at different locations on the worn surfaces.

<table>
<thead>
<tr>
<th>Cycles</th>
<th>Material</th>
<th>Ketac Molar</th>
<th>Ketac Molar C</th>
<th>FIXE</th>
<th>FIXEC</th>
</tr>
</thead>
<tbody>
<tr>
<td>(C-A) 50,000</td>
<td>18 (2)b</td>
<td>7 (2)b</td>
<td>22 (2)b</td>
<td>6 (2)b</td>
<td></td>
</tr>
<tr>
<td>(D-A) 200,000</td>
<td>83 (3)d</td>
<td>72 (4)d</td>
<td>93 (2)d</td>
<td>54 (5)d</td>
<td></td>
</tr>
<tr>
<td>recoat</td>
<td>recoat</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(C-A) 50,000</td>
<td>7 (1)a</td>
<td>-5 (15)a</td>
<td>9 (1)c</td>
<td>0 (3)a</td>
<td></td>
</tr>
<tr>
<td>(D-A) 200,000</td>
<td>57 (2)c</td>
<td>59 (6)c</td>
<td>70 (3)c</td>
<td>46 (4)c</td>
<td></td>
</tr>
</tbody>
</table>

Different lowercase letters indicate a statistically significant difference between the materials within the same experiment (in the same line) (p>0.01). Different uppercase letters indicate a statistically significant difference between the experiments (in the same column) within the same material (p<0.05).

Figure 5: Wear in µm per 50,000 and 200,000 cycles for the investigated materials at slip of 15% and 15 N force. The bars represent standard errors.
Discussion

The null hypothesis, stating that there was no difference in flexural strength between coated and uncoated specimens was rejected. Ketac Molar showed no difference in FS irrespective of the coat application but Fuji IX GP Extra showed statistically significant difference in FS depending on the use of the coat. Regarding the wear, the null-hypothesis was also rejected. There was a statistically significant difference in the use of the coat for both materials with all experiments, except for the one of Ketac Molar after 200,000 cycles.

Coating specimens with G-Coat Plus results in improved material properties in the laboratory. Despite some beneficial properties, the poor wear-resistance and the low fracture strength are still the most challenging properties of GICs, which limit their use to low-stress-bearing areas (22, 23). Using a coat might help to overcome these problems. Besides the coat, the long setting time can have a great influence on the sensitivity to water uptake (15). To avoid differences in this characteristic Fuji IX GP Extra was selected for this study as its setting time (2 min and 30 s) is the same as for Ketac Molar, which is much faster than the conventional Fuji IX GP (4 min and 30 s).

The wear rate of the uncoated specimens of Ketac Molar and Fuji IX GP Extra were consistent with those previously published (6). With exception of Ketac Molar after 200,000 cycles all groups showed significant differences between the coated and uncoated specimens. For all materials, with or without coat, a decrease in wear rate as function of time was observed. After 200,000 cycles the specimens showed a lower wear rate than after 50,000 cycles. The re-application of the G-Coat Plus reduced the wear rate for Fuji IX GP Extra after 200,000 cycles, but for Ketac Molar the wear rate was only lower in the short-term experiment. One can say that this might be due to the fact that the coating did not chemically interact with Ketac Molar but only superficially penetrated the material, forming an outer layer (Figure 3), which was totally removed in the longer wear experiment (200,000 cycles). This hypothesis is supported by Figure 4, in which the GIC detached from the coating despite the existence of micro-mechanical interlocking.

It has been shown that the three-point-bending test can represent the clinical situation (24), e.g. when the opposing cusp of the opposing tooth exerts forces onto the restoration. The ISO standard 9917-2 for dental water-based cements included only compressive strength as a mechanical property. The flexural strength is included in the ISO standard 9917-2 (for light-activated dental water-based cements). These standards were designed at a time when conventional GICs were not considered for use in occlusal and proximal cavities (25) and highly viscous GICs were not developed yet. However, an evaluation of the flexural strength seems to be more appropriate than compressive strength, as the bulk fracture is one of the main reasons for failure of proximal GIC restorations (25, 26). The coated surface in this experiment was placed opposite the applied force as it is the most stressed surface in which the crack will start. The results showed higher flexural strength compared to the previous version of those materials, the so-called low and medium viscosity GICs (27, 28). The flexural strength for these high viscosity versions of Ketac Molar and Fuji IX GP Extra are in line with previously reported values from hand mixed versions of those materials (29). While Fuji IX GP Extra showed significant improvement in its flexural strength with G-Coat, the same was not observed for Ketac Molar. Though Fuji IX GP Extra improved its flexural strength, this material seems to have lower strength than Ketac Molar. In the three-point-bending test the highest tension is on the opposite side of the applied force. Applying a coating at this side might prevent the existing flaws to serve as areas of stress concentra-
tion and crack propagation. Once a crack sets off, the specimen will fail due to its brittle nature. Apparently, the crack starts from within the specimen and not from the surface, meaning that the coat do not play a role when applied on materials which are intrinsically more resistant to flexural stresses. For some restorative materials, a correlation is observed between the surface damage and flexural strength (30). In the present study, we did not observe any correlation between the wear and the flexural strength, as the materials behaved differently for each property when the coat was used.

The manufacturer’s instructions recommend 20 s of light curing for G-Coat Plus. However, due to the length of the specimens (25 mm) in the FS experiment and the diameter of the curing light (10 mm), the curing was done in 3 times each 20 s along the bar. The coating was applied just after the first stage of the setting reaction. In the specimens of the wear test, it was re-applied after the first experiment. The aim of this re-application was to reproduce the clinical situation of coating the material when it is already for some time in the mouth, probably worn or which had already lost its coating layer. This was done to evaluate if the re-application of the coat can be done also in old restorations. According to our results, Fuji IX GP Extra showed significant improvement in wear-resistance and flexural strength when the G-Coat Plus was applied. Even after a long-wear experiment, when the coat was re-applied after 200,000 cycles, Fuji IX GP Extra with G-Coat Plus showed significantly better wear results than Fuji IX GP Extra without coat. This means that the coating can be used to protect the Fuji IX GP Extra restoration for at least 24 hours in the present experimental design. Recently, a research by Bagheri et al. (19) evaluated the fracture toughness of different esthetic restorative materials. Fuji IX GP Extra was either coated with G-Coat Plus or non-coated. The coated group showed significant differences compared to the non-coated group after 4 and 8 weeks of aging in water. Their results agree with the statement that water uptake in conventional GIC results in decrease in strength and elasticity of the material (31, 32). Another research by Schmage et al. (10) presented the wear rates for restorative composites, measured under similar circumstances. The values ranged between 18 and 50 µm after 200,000 cycles. Usually, restorative composites are required to have wear values lower than 50 µm per year (33). Fuji IX GP Extra with G-Coat Plus showed an average wear of 54 µm after 200,000 cycles, suggesting that the wear properties of these materials combined may approach the ones of composites.

Conclusions
G-coat Plus is indicated for concomitant use with Fuji IX GP Extra to decrease the early wear rate and increase its fracture strength. However, it should be considered that this study is limited to a short-term observation.
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References


