The ART of GIC proximal restorations in primary teeth

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Chapter 7

Micro-mechanical bond strength tests for the assessment of the adhesion of GIC to dentin

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**Abstract**

Aim: The aim of this study is to critically evaluate the bond strength (BS) of Glass-Ionomer Cements (GIC) to dentin with microtensile (μTBS) and microshear (μSBS) BS tests by assessing their rankings and failure patterns.

Methods: Samples were made on flat dentin surfaces and submitted to μTBS and μSBS. The materials tested were: high viscosity GIC (Ketac™ Molar Aplicap-KM), resin-modified GIC (Fuji II-FII), nano-filled resin-modified GIC (Ketac™ N100-N100) and an etch-and-rinse adhesive system with a composite resin (Adper™ Single Bond 2 and Z100™-Z100). All tests were performed with a Universal Testing Machine (24 h water storage, crosshead speed of 1 mm/min). Debonded surfaces were examined with a stereomicroscope (X40) to identify the failure mode. The data was analyzed with Two-way ANOVA (p<0.05) and LSD test.

Results: Means were statistically different regarding the tests and materials, indicating that values for BS obtained for each material depend on the test performed. Failure analysis revealed that failures produced by μTBS were mainly cohesive for KM and FII. μSBS failures were mainly adhesive or mixed for all materials. For the μTBS, the rank was Z100>FII>KM=N100, whereas for the μSBS it was Z100=FII=KM>N100.

Conclusion: It may be concluded that distinct micro-mechanical tests present different failure patterns and rankings depending on the material to be considered.
Introduction

Glass-Ionomer Cement (GIC) was introduced in the 70’s by Wilson and Kent (1) as a material that presented good marginal sealing properties. The characteristics of biocompatibility and fluoride release to adjacent structures, which might reduce secondary caries progression, as well as their less demanding clinical application technique and good retention in cervical lesions (2), characterize GICs as the material of choice for treatment of primary teeth in several countries. One of the GIC’s main advantages compared to other restorative materials is their chemical bond to enamel and dentin (3). It is well known that GIC bonds chemically to tooth surfaces (1, 4, 5). This chemical adhesion is formed through ionic and polar attractions between hydroxyapatite and polycarboxylate radicals in a way that the latter displace the phosphate and calcium ions from the former, which maintains the electrical neutrality (5) and promotes the chemical bond. This interaction layer is also considered to be beneficial in reducing the hydrolytic degradation, enhancing the restoration longevity (6).

The mechanical tests to assess bond strength of conventional GICs usually show cohesive failures within the material, indicating that the obtained values represent better the cohesive strength of the material itself rather than the interfacial bond strength between the material and the tooth (5). Because of that, some authors suggest that GIC bond strengths must be higher than the cohesive values frequently presented (1, 3). Different testing methods have been applied for bond strength measurements, such as tensile, shear, microtensile and microshear (7-9). The development of the bond micro-tests showed that smaller surface areas were associated with higher adhesive strengths, probably due to fewer flaws, as suggested by Griffith’s defect theory (7, 9, 10). Improvements were achieved with better attaching methods, delimitation techniques, and the micro-tests apparently showed a lower number of internal defects and more uniform interfacial stress distribution (7-12). Although both micro-tests are used to assess bonding effectiveness of GICs, it seems that the application of shear forces is more suitable for this kind of material due to its higher brittleness and fragility (13). Unfortunately, it remains unclear which test would be the most appropriate for evaluating the bond strength of GICs to dental hard tissues.

The purpose of this laboratory test was to critically evaluate the bond strength of GIC-based materials to dentin with microtensile and microshear bond strength tests by assessing their ranking and failure patterns. An etch-and-rinse adhesive and a composite were used as control material. The null hypotheses tested were that there is no difference in materials’ bond strength behavior (I) and in the failure patterns (II), irrespective of the test type used.

Material and methods

The study was approved by the research ethics committee of the School of Dentistry (University of São Paulo). Forty non-curious human third molars were stored up to one month in distilled water (5°C).

Experimental Design

The factors under study were mechanical tests, at two levels: microtensile and microshear, and adhesive restorative materials, at four levels: high viscosity GIC (Ketac™
Micro-mechanical bond strength tests for the assessment of the adhesion of GIC to dentin

Molar Aplicap), resin-modified GIC (Fuji II LC), resin-modified GIC with nanoparticles (Ketac™ N100) and etch and rinse adhesive system with a composite resin (Adper™ Single Bond 2 and Z100™). The experimental units were randomly divided into 8 groups according to the sets of variables forming a 4 x 2 factorial design (Figure 1).

![Figure 1: Diagram of experimental design.](image)

Specimen preparation

Each tooth was sectioned 3 mm above and below the cement-enamel junction (CEJ) using a slow speed diamond saw (Isomet 1000/Buehler Ltd., Lake Bluf, IL, USA). The occlusal side of the tooth section was ground with wet 120-grit silicon carbide sandpaper until the enamel was totally removed and the superficial dentin exposed. This procedure was controlled with aid of a stereomicroscope (SZ40 Olympus corporation, Tokyo, JP). Dentin surfaces were polished with wet 600-grit silicon carbide paper under running water for 1 min, for smear layer standardization.

All materials were applied in accordance with the manufacturers’ instructions. Samples of the Ketac™ Molar and Fuji II LC had their surfaces protected following the manufacturer instruction (Table 1).

For the light cured materials a halogen curing light was used (Jetlite 4000 Plus, J. Morita USA Inc., CA, USA) with 600 mW/cm², as confirmed by the device’s own radiometer. Prior to the bond strength measurements, all specimens were stored in distilled water at 37°C for 24 h.

Microtensile Bond Strength (µTBS)

Six tooth sections were selected (n=6) for each material. After the surface pre treatment, a crown was built over each surface using either one layer of Ketac™ Molar, or three to four layers of the other materials. The crown was up to a height of 5 mm.
The specimens were serially sectioned perpendicular to the bonding surface using a low speed diamond saw under water-cooling (Isomet 1000/Buehler Ltd., Lake Bluf, IL, USA) in order to obtain rectangular specimens of about 1 mm x 1 mm wide and 6-8 mm long (Figure 2). The final width and thickness of each specimen were measured with a digital micrometer (Mitutoyo Co, Tokyo, JP). The specimens were examined under X40 magnification using a stereomicroscope and the ones with visible interfacial defects were excluded.

The specimens were fixed to a Geraldeli jig (14) with cyanoacrylate glue (Loctite 454, Henkel Loctite Corp., Rocky Hill, CT, USA) and stressed at a crosshead speed of 1 mm/min until failure, using a universal testing machine (Mini Instron 4442 - Canton, MA, USA). The μTBS was expressed in MPa as derived from dividing the imposed force (N) at the time of fracture by the bond area (mm²). Premature failures were included for statistical analysis as 0 MPa.

Microshear Bond Strength (μSBS)

For each tooth surface, 3 specimens were made (Figure 2). Three lined up holes, 1 mm diameter, were punched in a double face adhesive tape (Tectape, Manaus, AM, BR), which was then positioned on the dentin surfaces (15). After surface pre treatment, a micro tube from a micro bore Tygon tubing (R-3603, Norton Performance Plastic, Cleveland, USA) with an internal diameter and height of 1 mm and 0.5 mm, respectively, was mounted over each hole of the tape. The materials were carefully inserted into the tubing lumens with a pediatric plunger (#0/1, Ice-Stainless, São Paulo, SP, BR). The specimens of Ketac™ Molar and Fuji II LC groups had their surfaces protected following the manufacturer’s instruction (Table 1). All specimens were stored in distilled water at 37ºC for 24 h before μSBS testing.

After removal of the tygon tubing with a scalpel, the specimen were checked under X40 magnification using a stereomicroscope for interfacial defects and the specimens showing formation of interfacial gap, bubble inclusion or any other defects were excluded from the study and replaced.

The teeth sections were adhered with cyanoacrylate glue (Loctite 454, Henkel Loctite Corp., Rocky Hill, CT, USA) to the testing apparatus, which in turn was placed in a universal testing machine (Mini Instron 4442, Canton, MA, USA) for μSBS testing. A thin steel wire (diameter 0.20 mm) was looped between the load cell projection and the restorative material cylinder, making contact through its lower half circumference and was gently held flush against the dentin/material interface (Figure 2P). A shear force was applied at 1 mm/min crosshead speed until the failure occurred (11). The fracture load (N) was recorded and the μSBS (MPa) was calculated.

Failure mode

All debonded surfaces were examined under a stereomicroscope at X40 magnification to identify the failure mode as: adhesive (apparently interfacial failure), cohesive in dentin, cohesive in material or mixed. Representative failed specimens were processed for field-emission scanning electron microscopy (FEI Quanta 600 FEG, Hillsboro, OR, USA) in order to illustrate the predominant failure mode in each group. The selected specimens were prepared using common specimen processing procedures, including fixation, dehydration, chemical drying, and platinum-sputter coating.
Table 1 – Materials used in this study. Dentin pre-treatment and restoration surface protection.

<table>
<thead>
<tr>
<th>Material</th>
<th>Composition</th>
<th>Application</th>
<th>Surface Protection</th>
</tr>
</thead>
<tbody>
<tr>
<td>(KM) Ketac™ Molar Aplicap™ (3M ESPE, Seefeld, Germany)</td>
<td>Ketac™ Conditioner: polyacrylic acid (25%) Powder: calcium aluminum-lanthanum-fluorosilicate glass, acrylic acid-maleic acid copolymer, pigments Liquid: water, acrylic acid-maleic acid copolymer, tartaric acid</td>
<td>Apply Ketac Conditioner (10 s); rinse with copious amount of water; gently air-dry (5 s); leaving a moist surface. Activate the capsule for 2 s; automatically mix the capsules (15 s); apply to enamel and dentin surfaces.</td>
<td>Ketac™ Glaze</td>
</tr>
<tr>
<td>(FII) Fuji II LC (G. C. Europe, Leuven, Belgium)</td>
<td>Cavity conditioner: 3% aluminum chloride, 20% polyacrylic acid Powder – fluoroalumino-silicate glass Liquid – polyalkenoic acid, HEMA, dimethacrylate, camphorquinone, water</td>
<td>Apply cavity conditioner and leave undisturbed (10 s); rinse with water (10 s); gently air-dry (5 s); leaving a moist surface. Automatically mix the capsules (10 s); apply to enamel and dentin surfaces; light-cure for 20 s.</td>
<td>G-Coat PLUS</td>
</tr>
<tr>
<td>(N100) Ketac™ N100 Nano Light-Curing (3M ESPE, Seefeld, Germany)</td>
<td>Primer: water (40-50%); HEMA (35-45%); acrylic/itaconic acid copolymer (10-15%); photoinitiators. Ketac™ N100 Nano : De-ionized water, HEMA, vitrebond copolymer \9 a methacrylate modified polyalkenoic acid), florualuminosilicate glass, nanomers and nanoclusters.</td>
<td>Dispense and apply the nano-ionomer primer during 15 s; air-dry for 10 s (shiny surface); light cure (10 s); Dispense 2 clicks of the Ketac N100; mix for 20 s (uniform color); apply incrementally (&lt;2mm); light-cure each layer for 20 s.</td>
<td>Not recommended</td>
</tr>
<tr>
<td>Z100™ (3M ESPE, Seefeld, Germany) – used with Adper™ Single bond 2.</td>
<td>Bis-GMA, TEGDMA, zirconia, silica.</td>
<td>Etch (Phosphoric acid 35% - 15 s); rinse with water (10 s); gently air-dry (5 s); light-cure (10 s). Insert the composite incrementally (&lt;2mm); light-cure each layer for 40 s.</td>
<td>Not recommended</td>
</tr>
</tbody>
</table>

Statistical analysis
For an analysis of the different tests (inter-method), the percentage of the different failure modes were calculated for each material and test method. A chi-square test was performed to analyze if the failure mode of each material was test-dependant. For quantitative analysis, two-way ANOVA and LSD (Least Square Difference) post hoc test were used to examine differences in the calculated bond strengths (MPa) between the different tests and materials. Due to the absence of homocedasticity, it was necessary to transform the
bond strength data into logarithmic data. A p-value lower than 0.05 was taken as statistically significant for all tests. We calculated the sample power. Considering the effect size of 0.8 (large), the power was 0.88, which represent a reliable sample for detecting differences between groups.

Results
Comparing the four materials submitted to μTBS, significant differences (p<0.001) were found among the materials. Nevertheless, Ketac™ Molar and Ketac™ N100 were statistically similar and presented significantly lower bond strength values than Fuji II LC, which was, on its turn, significantly lower than for Z100™. For μSBS, Ketac™ Molar, Fuji II LC and Z100™ showed no difference among each other and presented significantly (p<0.001) higher values than Ketac™ N100.

The bond strengths for all materials were ranked as: Z100>FII>KM=N100 for the μTBS; whereas for the μSBS it was Z100=FII=KM>N100. The descriptive results from failure analysis are graphically summarized in Figure 3. A great number of specimens submitted to μTBS, tended to fail cohesively in the material while the μSBS led to a majority of adhesive and mixed failures (Figure 4). The chi-square test revealed a significant test-dependence on the failure modes for KM (p=0.0002) and FII (p<0.001). The microtensile test showed a higher number of cohesive (in material/dentin) and mixed failures, while the microshear test produced a majority of adhesive failures. For N100 (p=0.055) and Z100 (p=0.43) this pattern was not observed, as there was no difference in failure modes between the tests (the two materials had a large count of both adhesive and mixed failures).

ANOVA revealed an interaction effect between test and material (p<0.001). Table 2 shows the mean and standard deviation values for bond strength of all materials. The two different bond strength tests produced significantly different (5%) bond strength values in all four materials.

Discussion
Even presenting poorer physical-mechanical properties when compared to composites, GICs have been demonstrating an excellent clinical retention rate in cervical lesions (3, 16) and in ART single-surface cavities (17). Therefore, the low laboratory bond strength values may not correspond to the clinical outcomes (6, 8, 17). Microtensile and microshear bond strength tests and the qualitative analysis of the failure mode in this study were performed to give a better elucidation of the GIC’s behavior regarding the different bond strength micro-tests, and thus possibly establish some relation to the clinical findings.

The results found in the present study showed that the bond strength means were statistically different regarding the tests and materials. This indicates that bond strength values of the materials depend on the test method. This is in line with other papers, which investigated the bonding mechanism with adhesive systems (18, 19). To the best of our knowledge, this is the first paper that evaluated the bonding test effect with GIC.
Figure 2: Schematic illustration of the $\mu$TBS and $\mu$SBS specimen preparation.
The bond strength values found for Ketac™ Molar were significantly higher in μSBS than in μTBS, whereas for all other materials, this pattern was not repeated. The failure modes of KM were significantly test dependant, with a predominance of adhesive and mixed failures for μSBS, whereas failures were mainly cohesive for μTBS. This indicates that μSBS was possibly more appropriate to evaluate the bond strength instead of the cohesive strength of this material. This greater number of cohesive failures found for μTBS can explain the lower values found for KM in such test as they are more related to a cohesive failure or to a tensile strength than to the bond strength itself. For this specific material the tensile strength (20) seems to be lower than the bond strength, which might explain the lower values for KM in μTBS.

The failure mode for Fuji II LC was found to be 91.67% adhesive for μSBS, as for μTBS only 15.57% was adhesive. The chi-square test revealed the failure mode was significantly test dependent (p<0.001). Moreover, the mean averages for μTBS were significantly higher. Therefore, again, this might also reinforce the fact that the μTBS test measured mostly the tensile strength rather than the bond strength. This would explain the higher values found for μTBS, since the tensile for RMGIC is higher than for high viscosity GIC (20).

The nano-RMGIC (Ketac™ N100) presented the lowest bond strength values. The failure analysis showed a similar behavior in both tests with mostly adhesive failures. Other authors observed mainly adhesive failures between the nano-RMGIC and dentin (21, 22). Investigating the shear bond strength (SBS) of different nano-filled materials to dentin, Korkmaz et al. (22) found the lowest bond strength values for the nano-filled RMGIC. The μTBS results from Coutinho et al. (21) showed that the nano-filled RMGIC bonded as effectively to dentin as a conventional GIC, but less effectively than the other RMGIC (Fuji II LC), which are in accordance to our μTBS results.

The control group (composite resin) presented higher mean values for μTBS than μSBS. The failure patterns were similar in both tests, with mostly adhesive and mixed failures.

We should be careful when comparing nominal values from both tests as the difference can be attributed to methods, heterogeneous stress distribution at the interface and also to the occurrence of cohesive failures (11, 23). Presenting higher bond strength in one specific test does not mean a greater degree of reliability. The ranking of materials was different for the two tests. It was Z100>FII>KM=N100 for μTBS; whereas for μSBS

### Table 2 – Bond Strength means (MPa) and standard deviations (SD). Different uppercase letters indicate a statistically significant difference (p<0.05) for each material in the two different tests (horizontal lines). Different lowercase letters indicate a statistically significant difference between the materials within each test (vertical lines) (p<0.001).

<table>
<thead>
<tr>
<th>Material</th>
<th>μTBS - Mean (SD)</th>
<th>μSBS - Mean (SD)</th>
</tr>
</thead>
<tbody>
<tr>
<td>KM</td>
<td>9.5 (1.9) ^A,a</td>
<td>17.2 (3.0) ^B,b</td>
</tr>
<tr>
<td>FII</td>
<td>23.8 (4.7) ^B,b</td>
<td>18.3 (2.8) ^A,b</td>
</tr>
<tr>
<td>N100</td>
<td>9.4 (3.8) ^B,a</td>
<td>6.8 (1.4) ^A,a</td>
</tr>
<tr>
<td>Z100</td>
<td>50.7 (14.5) ^B,c</td>
<td>21.0 (5.5) ^A,b</td>
</tr>
</tbody>
</table>
Comparing rankings or methods is also questionable as there is a range of differences that should be taken into account in the analysis, including all the factors quoted above and mainly the operator running the tests (8, 9, 23). The ranking presented by the µSBS test seems to be more related to the outcome of clinical studies as far as GICs are taken into account (24).

An advantage of the µSBS test is the absence of sectioning, trimming and shaping procedures after bonding the specimen. Such treatments possibly affect the results as they can create micro-cracks in the material, which might become areas of stress concentration that may propagate the crack during mechanical loading (9, 11). The mold removal is the only pre-stress caused to the specimen prior to its testing (11). The loading method, with the wire correctly positioned in the interface area, can help preventing premature failure (8). Therefore, for brittle materials such as the GIC, µSBS seems to be better indicated when compared to µTBS, as their low cohesive strength predictably leads to cohesive failures when they are tested in tension.
Bond strength variation for the different materials can be related to adhesion mechanisms and also to the individual material’s composition (25). The adhesion mechanisms of GIC to the tooth structure are attributed to ionic and polar links between the carboxylic groups and the tooth surface (4). These interactions are initiated by a weak hydrogen bond, and it is gradually strengthened by a polar/ionic interaction. Then, polyalkenoic acid chains will spread in the enamel and dentin, dislocating calcium and phosphate ions from the apatite crystals (25). Another bond mechanism attributed to GIC is its micromechanical retention, established by the diffusion of GIC components within the collagens fibers exposed by the polyacrylic acid, thus forming a layer similar to the hybrid layer (5).

Figure 4: SEM micrographs of µSBS (a,b,c,d) and µTBS (e,f,g,h) debonded surfaces. (a) Ketac Molar mixed failure (b) Fuji II LC adhesive failure (c) Ketac N100 adhesive failure (d) Z100 mixed failure, (A) adhesive and (S) cohesive in substrate, arrows indicate some material remaining in the surface. (e) Ketac Molar cohesive failure (f) Fuji II LC adhesive failure (g) Ketac N100 mixed failure (C) cohesive failure (A) adhesive failure (h) Z100 mixed failure, (C) cohesive failure within the material (S) cohesive failure within the substrate.

The dentin pre treatment can also influence the bonding mechanism. Polyacrylic acid is indicated to increase the bond efficiency of GIC as it creates a superficial cleaning and a partial demineralization, which enhances the possibilities of chemical and micromechanical interaction with the hydroxyapatite (5). Figure 5 shows smear layer remaining on the dentin surface of a sample prepared with Ketac™ N100 after debonding, which can contribute to an insufficient adaptation and to a weaker adhesion. Based on the failure mode and the ranking of bond strength values of each test, we conclude that different materials behave differently depending on the bond strength micro-test employed. Altogether, the bonding effectiveness of GIC-based materials may be more dependent on the characteristics of the mechanical test used than on their actual interaction with the substrate. This may explain the incongruence between clinical and laboratory results as far as the adhesion of GIC-based materials is concerned. It may also explain why in our in vitro test the bonded composite performed better than any of the tested GIC, while clinical evaluations usually favor GIC when it comes to bond performance. Considering these
findings, in vitro bond strength values and their clinical performance may not be well correlated.

![N100 micrograph showing the smear layer (*) remaining in a debonded specimen.](image)

**Figure 5:** N100 micrograph showing the smear layer (*) remaining in a debonded specimen.

**Acknowledgments**

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