Bond Strength Evaluation of Three Self-adhesive Luting Systems Used for Cementing Composite and Porcelain

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Clinical Relevance
Although all self-adhesive systems give improved handling and ease of use compared to traditional multi-step adhesive cements, clinicians should still consider self-adhesive cements as a heterogeneous category of luting agents that need to be better classified in terms of bond strength and chemical/mechanical properties.

SUMMARY
Self-adhesive resin cements were recently introduced with the purpose of simplifying the cementation technique, as they combine the use of adhesive and cement in a single application, eliminating the need for pretreatment of the tooth. In the present study a microtensile bond strength test (μ-TBS) was used to compare three self-adhesives, an etch-and-rinse and a self-etch luting system, in the cementation of resin-based composite (RBC) and ceramic disks to dentin. Freshly extracted molars were transversally sectioned to expose flat, deep dentin surfaces. Cylindrical specimens (5 mm in diameter and 10 mm in height), consisting of RBC disks and leucite-based glass ceramic disks, were produced. The RBC disks were sandblasted with 50-μm Al₂O₃. The ceramic disks were conditioned with 9.5% hydro-
fluloric acid gel and silane application. All of the disks were then bonded to dentin surfaces employing five different luting agents: iCEM Self Adhesive (Heraeus Kulzer), MaxCem (Kerr Corporation), RelyX UniCem (3M ESPE), EnaCem HF (Micerium), and Panavia F2.0 (Kuraray-Dental). The products were applied according to the manufacturers' instructions. The specimens were sectioned perpendicular to the adhesive interface to produce multiple beams measuring approximately 1 mm² in cross section. For each experimental group 12 beams were tested. The preterm failures were also taken into account. All of the specimen preparations were performed by the same operator. The beams were tested under tension at a crosshead speed of 0.5 mm/min until failure. Mean μ-TBS values were calculated for each group. Data were analyzed by a two-way analysis of variance, and multiple comparisons were performed using a Tukey test (α=0.05). The UniCem group showed the lowest number of preterm failures among the tested self-adhesive systems. When premature debondings were included in the mean value calculation, bond strength values for the UniCem group were statistically equal to or even higher than those achieved with the other self-adhesives, although these values were still statistically worse than those obtained using traditional multi-step luting agents.

INTRODUCTION

Resin-based composite (RBC) and ceramic indirect restorations represent an up-to-date and valid therapeutic option that is increasingly adopted clinically. They provide optimal esthetics, ensuring high patient satisfaction and mechanical properties that give good functional results. Posterior ceramic bonded partial restorations are a conservative and esthetic approach for compromised teeth. Overlays constitute a less invasive alternative for tooth tissues than do crown preparations. Partial restorations are also indicated in cases of full arch or quadrant rehabilitations including several teeth. They are routinely cemented on prepared dental surfaces using adhesive techniques.5-4

Traditional adhesive systems promote bonding by creating a hybrid layer on acid-etched dentinal surfaces through an amphiphilic primer infiltration into the conditioned substrates.5-7 Among the different resin cements available on the market, the recently introduced self-adhesive resin systems are increasing in popularity, as they contain both adhesive and cement in one single formulation, promoting simultaneous demineralization and penetration without the need for etching and bonding. The simplification of this critical phase into one single application renders these products less operator sensitive compared to traditional resin cements.8-13 Many studies have evaluated the bond strength and behavior of self-adhesive resin cements, comparing them to traditional multi-step techniques.

Different self-adhesive resin cements that are commercially available vary in their composition and physical properties. To date few studies exist that compare different self-adhesive resin cements employed to lute composite and porcelain. The aim of this study was to compare the microtensile bond strength (μ-TBS) of resin composite and ceramic indirect restorations cemented to dentin using three different self-adhesives, an etch-and-rinse and a self-etch luting system. The null hypothesis was that there would be no significant difference among the resin cements applied.

MATERIALS AND METHODS

Tooth Preparation

The experimental procedures performed in this study are summarized in Figure 1. Recently extracted molars that were free of fractures, caries, and restoration were used. Remaining debris was removed with an ultrasonic scaler. Teeth were rinsed and stored in 0.5% Chloramine T at 4°C for not more than three months. To expose flat dentin surfaces, the crown was cut perpendicularly to the long axis at approximately 3 mm from the cemento-enamel junction with a low-speed diamond saw (Micromet M, Remet, Casalecchio di Reno, Bologna, Italy) under copious water. The flat surfaces were further ground using a 180-grit silicon carbide (SiC) paper under running water for 30 seconds to obtain a standardized smear layer. A stereomicroscope (Nikon SMZ10; Tokyo, Japan) was used to ascertain the complete absence of any enamel residue. If needed, the surfaces were further worn until the complete removal of enamel was achieved.

Microhybrid RBC (Enamel-Plus HFO UD3; Micerium, Avegno, Genova, Italy) cylinders were manufactured from transparent polyethylene molds measuring 5 mm in diameter and 10 mm in height. The mold was positioned on a glass surface and then filled with the RBC. Resin composite was applied in five layers with a 2-mm thickness. Each layer was
individually polymerized for 40 seconds (L.E. Demetron I; Sybron/Kerr, Orange, CA, USA) with a 1200-mW/cm$^2$ output). After mold removal, cylinders underwent a further heat-curing cycle in an oven (Bulb PlusT; Micerium) at 70°C for 10 minutes. All composite surfaces were then ground with 600-grit SiC paper and further roughened using an intraoral air-abrasion device (Micerium) with 50 l mA l$^2$O$^3$ particles (Korox, Bego, Bremen, Germany). The tip of the sandblaster was held 5 mm from the composite surface for 10 seconds at 2-bar pressure. Each sample was then washed under running water and placed in an ultrasonic bath to remove any debris.

Ceramic blocks (IPS e.Max; Ivoclar Vivadent, Solna, Sweden) were sectioned in smaller cylinders of approximately 5 mm in diameter and 10 mm in width and were further ground with 400-grit and 600-grit SiC paper under running water until smooth surfaces were obtained. Hydrofluoric acid (IPS Ceramic etching gel; Ivoclar Vivadent) was applied on the adhesion surface for 60 seconds, then thoroughly rinsed with water and air-dried. Next, silane (Porcelain Prep Kit, Pulpdent) was applied and left undisturbed for 60 seconds, after which the surface was again air-dried.

**Bonding Procedures**

Teeth were subdivided into five groups (iCEM, MaxCem, UniCem, EnaCem, and PanF2.0) according to the resin cement employed (Table 1).

For the iCEM, MaxCem, and UniCem groups, no acidic pretreatments were performed and no adhesive was applied on the tooth surfaces; specimens were simply cleaned with an air-water syringe and dried with cotton buds. Subsequently the cement was applied.

For the EnaCem group, the dentin surfaces were first etched for 15 seconds with a 36% phosphoric acid gel and were then thoroughly washed using a water spray for at least 15 seconds. The excess water was blot-dried from the dentin surface with a wet cotton pellet, leaving the surface visibly moist. An equal number of drops of the bonding agent and its activator (Table 1) were mixed in a mixing well for two seconds. Generous amounts of mixed adhesive/activator were rubbed onto the moist dentin with a microbrush (Microbrush X; Microbrush Corp, Grafton, WI, USA) for 10 seconds and air-thinned with two to three short, moderate blasts of air. Equal amounts of the dual-cure self-activating system...
base paste and catalyst were mixed and applied to the flat dentin surfaces. For the PanF2.0 group, no acid-etching was performed, according to the manufacturer’s instructions. An equal number of drops of ED Primer II liquids A and B (Table 1) were mixed, rubbed onto the dentin surface, left in place for 30 seconds, then air-thinned with two to three short, moderate blasts of air. Panavia F2.0 paste A and B (Table 1) were mixed in equal amounts and placed over the dentin surfaces.

After cement application, composite and ceramic cylinders were positioned on the dentin surface. The bonded assemblies were held centrally between the two measuring arms of the vertically positioned digital micrometer. A load pressure of about 5 N was applied on the cylindrical RBC or ceramic specimens in order to standardize and to simulate the clinical conditions of inlay cementation. This pressure was repeated three times for five seconds each time at intervals of 15 seconds. The luting cement thickness was kept at approximately 100 \( \mu m \). The micrometer arms were slowly adjusted to produce a reading that was 100 \( \mu m \) (mean) thicker than that initially recorded for the respective dentin specimen and RBC/ceramic inlay. Excess cement was removed with a pointed (sharp) instrument before complete polymerization. After the initial auto-polymerization a further photo-polymerization was conducted under a load of 5 N using an LED light (SmartLite PS; Dentsply DeTrey, Konstanz, Germany; output: 950 mW/cm\(^2\)) from five different directions for a total exposure time of 200 seconds. After 24 hours in distilled water at 37°C, all samples underwent 5000
thermal cycles in deionized water from 5°C to 55°C with 30 seconds of dwell time and five seconds of transfer time between baths.

Samples were then sectioned perpendicularly to the adhesive interface with a diamond saw (Micromet M; Remet, Casalecchio di Reno) under continuous running water, which acted as coolant and lubricant, obtaining beams with an adhesive surface of approximately 1 mm². Four samples of each tooth’s central part were collected. All of the bonding procedures and specimen preparations were performed by the same expert operator.

**Microtensile Bond Strength Test**

Specimens were secured to the arms of a Universal Testing Machine (LR30K; Lloyd Instruments Ltd, Fareham, UK) by an anchoring device and using cyanoacrylate and were then subjected to traction at a crosshead speed of 0.5 mm/min until fracture. The anchoring device was interposed with a chain with two rings connected to an upper clamp. Specimens were then removed, and the cross-sectional area of the fracture sites was measured with a digital caliper (series 500 Caliper; Mitutoyo America Corp, Aurora, IL, USA) to calculate the ultimate tensile bond strength expressed in MPa. Specimen preparation and μ-TBS procedures were carried out until 12 measurements were obtained for each combination of the five luting agents with the two substrates under investigation (n = 12). Each beam that underwent spontaneous interfacial debonding throughout specimen cutting and anchoring to the testing device procedures was considered as a preterm failure; the number of the premature failed beams observed in each group was recorded.

**Mode of Failure**

After the μ-TBS test, both the dentin and RBC/ceramic sides of the fractured beams were mounted on aluminum stubs, gold–sputter-coated, and observed by scanning electron microscope (SEM; EVO MA 15; Carl Zeiss NTS GmbH, Oberkochen, Germany) at 190× or higher magnification for fracture mode determination. The failure modes were classified into one of six different types, as follows:

- Type 1: Cohesive failure in dentin;
- Type 2: Adhesive failure at the luting-dentin interface;
- Type 3: Mixed adhesive failure and cohesive failure in dentin;
- Type 4: Cohesive failure in the luting agent;
- Type 5: Mixed adhesive failure and cohesive failure in RBC (or ceramic); and
- Type 6: Adhesive failure at the luting-RBC (or ceramic) interface.

**Data Analysis**

Data were arranged on the basis of the material employed (Table 1) and the adhesive substrate (RBC or ceramic) and were then processed according to three different methodologies proposed by previous studies. In a first attempt, although the number of preterm failed beams was recorded, they were not considered for mean value computation. Subsequently mean values were calculated by attributing to each lost beam an arbitrary value that was half of the lowest μ-TBS value recorded in the present study. Finally, a zero value was assigned to each preterm failed sample. Statistical analysis was performed using SPSS Advanced Statistical 11.5 software for Windows (SPSS, Chicago, IL, USA). A two-way analysis of variance was used to evaluate the influence of cement type and the substrate on the bond strength values. Multiple comparison analysis using the Tukey test was carried out. Values of p lower than 0.05 were considered statistically significant in all tests.

**RESULTS**

The mean μ-TBS values and numbers of preterm failures observed in the experimental groups are summarized in Tables 2 through 4. Table 2 shows the mean values calculated without taking into account the number of preterm failed beams. Using this data processing methodology, MaxCem showed statistically higher values on ceramic substrates compared with all the other groups, which did not differ from one another. On RBC the highest values were recorded with etch-and-rinse and self-etch systems, while among the self-adhesives, UniCem achieved the best results, and they did not differ from those obtained with the same cement on ceramic substrates. When an arbitrary value (Table 3) or a zero value (Table 4) was assigned to each preterm lost beam, UniCem yielded higher mean μ-TBS values than did MaxCem and iCEM, but lower values than EnaCem and PanF2.0. The differences were statistically significant only among RBC groups.

On both substrates, UniCem showed a number of preterm failures that was comparable with those observed in EnaCem and PanF2.0 groups and lower than in the MaxCem and iCEM groups.
SEM analysis of the specimens revealed a prevalence of adhesive failures at the luting-substrate interface (type 6) for all ceramic groups, with few differences among the luting agents. Concerning the RBC groups, the prevalent fracture pattern was an adhesive failure at the luting-dentin interface (type 2) for PanF2.0 and for the three self-adhesive groups and a cohesive failure in the luting agent (type 4) for EnaCem group.

**DISCUSSION**

In the present study the three self-adhesive systems investigated demonstrated different abilities with regard to the cementation to dentin of both resin composite and ceramic substrates.

In Table 2 the mean μ-TBS values registered in the experimental groups, not taking into account the number of preterm failed beams, are shown. These values have to be considered to be overestimated. Although on ceramic substrate, the MaxCem values shown in Table 2 were statistically indicated to be the highest, when preterm lost beams were considered for mean value calculation, assigning them an arbitrary value (Table 3) or a zero value (Table 4), statistical significance dramatically changed, indicating UniCem as the luting agent able to yield the strongest bond strength. This indicates that the role of preterm failures should not be neglected for a proper interpretation of μ-TBS results. This is in accordance with the methodology of recent μ-TBS studies17-19 that kept the amount of premature

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### Table 2: Means (and Standard Deviations, SDs) of the Microtensile Bond Strength Test (μ-TBS) Values Registered in the Experimental Groups, Not Including Preterm Failed Beams into the Calculation

<table>
<thead>
<tr>
<th>μ-TBS, MPa (SD)</th>
<th>Luting Agent</th>
<th>ICEM</th>
<th>MaxCem</th>
<th>UniCem</th>
<th>EnaCem</th>
<th>PanF2.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>Substrate IPS e.Max Ceramic</td>
<td>10.23 α₁ (3.77)</td>
<td>15.37 α₁ (2.60)</td>
<td>9.74 α₂ (3.99)</td>
<td>9.46 α₂ (2.20)</td>
<td>8.88 β₂ (3.02)</td>
<td></td>
</tr>
<tr>
<td>Lost/tested beams</td>
<td>76/12</td>
<td>48/12</td>
<td>20/12</td>
<td>12/12</td>
<td>19/12</td>
<td></td>
</tr>
<tr>
<td>Enamel Plus HFO RBC</td>
<td>6.46 α₁,α₂ (2.48)</td>
<td>4.15 α₂ (2.24)</td>
<td>9.44 α₁ (5.15)</td>
<td>30.37 α₁ (9.98)</td>
<td>17.82 β₁ (6.02)</td>
<td></td>
</tr>
<tr>
<td>Lost/tested beams</td>
<td>8/12</td>
<td>8/12</td>
<td>0/12</td>
<td>0/12</td>
<td>0/12</td>
<td></td>
</tr>
</tbody>
</table>

Abbreviation: RBC, resin-based composite. 

α Same online small-capital letters indicate no differences among different levels of the factor “Luting Agent” (reading horizontally). 

### Table 3: Means (and Standard Deviations, SDs) of the Microtensile Bond Strength Test (μ-TBS) Values Obtained in the Experimental Groups, Attributing to Each Lost Beam Half of the Lowest Value Observed in the Present Study

<table>
<thead>
<tr>
<th>μ-TBS, MPa (SD)</th>
<th>Luting Agent</th>
<th>ICEM</th>
<th>MaxCem</th>
<th>UniCem</th>
<th>EnaCem</th>
<th>PanF2.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>Substrate IPS e.Max Ceramic</td>
<td>2.38 α₁ (3.41)</td>
<td>3.98 α₁ (5.85)</td>
<td>4.47 α₂ (4.90)</td>
<td>5.30 α₂ (4.52)</td>
<td>4.13 α₂ (4.25)</td>
<td></td>
</tr>
<tr>
<td>Lost/tested beams</td>
<td>76/12</td>
<td>48/12</td>
<td>20/12</td>
<td>12/12</td>
<td>19/12</td>
<td></td>
</tr>
<tr>
<td>Enamel Plus HFO RBC</td>
<td>4.11 α₁ (3.49)</td>
<td>2.73 α₁ (2.46)</td>
<td>9.44 α₁ (5.15)</td>
<td>30.37 α₁ (9.98)</td>
<td>17.82 β₁ (6.02)</td>
<td></td>
</tr>
<tr>
<td>Lost/tested beams</td>
<td>8/12</td>
<td>8/12</td>
<td>0/12</td>
<td>0/12</td>
<td>0/12</td>
<td></td>
</tr>
</tbody>
</table>

Abbreviation: RBC, resin-based composite. 

α Same online small-capital letters indicate no differences among different levels of the factor “Luting Agent” (reading horizontally). 

β Same numbers indicate no differences among different levels of the factor “Substrate” (reading vertically).
debondings in consideration, as it might give an important predictive indication of the adhesive effectiveness.

On ceramic substrate, the number of preterm failures observed with UniCem was slightly higher than that observed with the traditional systems (EnaCem HF and Panavia F2.0), but visibly lower than those associated with the other self-adhesives, MaxCem and iCEM. Furthermore, no preterm failures were recorded when UniCem was used on RBC. This finding, together with the relatively high μ-TBS values observed (Tables 3 and 4), might indicate the greater reliability of UniCem compared to the other self-adhesive luting agents under investigation.

An overestimation of the mean values may also justify the overall higher bond strengths showed for porcelain compared to RBC groups in Table 2; a similar finding is observed just for MaxCem in Tables 3 and 4, although these values do not achieve statistical significance.

In the present study, the best bonding performances were achieved with the traditional etch-and-rinse and self-etch systems.

In brief, the adhesive mechanism of multi-step etch-and-rinse systems involves a phosphoric acid-etch step that within enamel produces deep etch-pits in the hydroxyapatite-rich substrate and within dentin demineralizes up to a depth of a few micrometers to expose a hydroxyapatite-deprived collagen mesh. The next step involves either a separate priming step followed by the application/curing of a combined primer/adhesive resin following a simplified two-step procedure or a separate primer and adhesive resin step following a three-step procedure. The final objective is to micromechanically interlock upon diffusion and in situ polymerization of monomers into the enamel etch-pits, the opened dentin tubules, and the exposed collagen network, the latter forming the well-documented hybrid layer.

The self-etch approach of Panavia F2.0 is based upon the dissolution of the smear layer, without the subsequent removal of the dissolved calcium phosphates, as there is no rinse phase. Functional monomers like 10–methacryloyloxydecyl dihydrogen phosphate have been proven to interact with this residual hydroxyapatite through primary ionic binding. The resultant twofold micromechanical and chemical bonding mechanism closely resembles that of glass ionomers.

On the contrary, the basic adhesion mechanism of the different self-adhesive cements is only partially understood; multifunctional methacrylated phosphoric ester monomers are considered able to etch dental tissue as a result of initial acidity and to simultaneously infiltrate demineralized enamel and dentin substrate. The subsequent setting reaction occurs through the radical polymerization of alkaline filler particles reacting with acidic monomers, leading to a pH increase. The bond obtained is termed micromechanical, as the chemical interaction between Ca ions derived from hydroxyapatite and the functional monomers might aid in the infiltration of monomers into etched dental tissue; however, little is known about this chemical interaction.

Differences in terms of adhesion among materials that undergo similar reactions could be due to their
composition. The investigated luting agents differ by the type of functional monomer, percentage of inorganic filler, and pH. Monomers interact similarly with the dental substrate as determined by different initial pH values. The initial system acidity provokes suitable etching of tissue, enabling an optimal micromechanical bond. Although an initial low pH value is desirable for better etching of mineralized teeth structure, its persistence can interfere with an effective adhesion.25,26

Both MaxCem and UniCem gave low pH values (2.0 and 2.8, respectively) 90 seconds after mixing, ensuring a sufficient initial acidity. However, after 48 hours, only UniCem recorded a neutral pH, while MaxCem still presented an acidic (2.4) pH value.26

Mazzitelli and others27 assumed that if the acidic monomers are not properly neutralized they might retain their etching potential, affecting the polymerizing reaction and jeopardizing adhesion. UniCem has a 72% weight filler load compared with 66% for MaxCem. The filler content of iCEM is even lower (46% wt). Differences in acidity might be related to the presence of different functional monomers and filler levels as they contribute to the initial pH.26 Moreover, a higher filler percentage in resin cements is considered responsible for providing better mechanical properties,26 and so the higher bond strength for UniCem could be attributed to a higher fraction of inorganic components.

Vrochari and others28 found that RelyX UniCem was able to comply with the International Standards Organization requirements regarding water sorption and solubility, while MaxCem was not, as it yielded very high sorption values and the greater mass loss. This might support the findings of the present study, as it has been shown that among one-step and self-etching systems, the more hydrophilic ones tend to show lower bond strengths29 and reduced tensile strengths.30

Our results are in accordance with those of Behr and others,31 who highlighted the better performances of UniCem, with regard to marginal adaptation, compared to MaxCem and to a third self-adhesive luting agent, following ceramic disk cementation to dentin. Marginal analysis has been reported to provide indications of adhesive system ability to compensate for shrinkage of resins during polymerization.32 The higher bond strength for UniCem may also derive from its greater ability to chemically bond with hydroxyapatite.23

Many authors have discussed the actual ability of self-adhesive luting agents to determine a tangible infiltration of the dental surface,9,33 compared to traditional systems. Recent studies that did not consider premature debonding showed UniCem achieving high bond strengths for ceramic14 and good values for composite cementation,14,15 which is in accordance with the results summarized in Table 2, even though etch-and-rinse adhesive systems were still reported to be more reliable on RBC substrate.14,15 Traditional adhesive systems may lead to a better demineralization of dentin with the maximum smear layer removal, exposing a greater quantity of dentinal tubules and collagen fibers to resin infiltration by hydrophilic primer, thus clearly yielding a stronger and more effective bond between resin and the substrate.

CONCLUSIONS

Within the limitations of an in vitro study, the present findings showed that just one among the self-adhesive systems investigated was able to produce predictable adhesion in terms of bond strength and number of preterm failures on both ceramic and RBC substrates. As a consequence, self-adhesive luting agents still should be considered a heterogeneous group of resin cements with substantial differences among them in terms of setting reaction, chemical composition, and pH. Further studies seem required to broaden the existing knowledge about the chemical and mechanical properties that could clarify the different performances recorded and might lead to a more specific classification of these newly marketed materials.

Disclosure

The authors declare that they have no conflict of interest and that they didn’t receive material supplies or financial supports by any of the manufacturer industries.

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