Bonding effectiveness of adhesive luting agents to enamel and dentin

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ABSTRACT

Objectives: The bonding effectiveness of five adhesive luting agents to enamel and dentin using different application procedures was determined using a micro-tensile bond strength protocol (μTBS).

Methods: Enamel/dentin surfaces of human third molars were flattened using a high-speed diamond bur. Composite resin blocks (Paradigm, 3M ESPE) were luted using either Linkmax (LM; GC), Nexus 2 (NX; Kerr), Panavia F (PN; Kuraray), RelyX Unicem (UN; 3M ESPE) or Variolink II (VL; Ivoclar-Vivadent), strictly following manufacturers’ instructions. For some luting agents, modified application procedures were also tested, resulting in four other experimental groups: Prompt L-Pop + RelyX Unicem (PLP + UN; 3M ESPE), Scotchbond Etchant + RelyX Unicem (SE + UN; 3M ESPE), Optibond Solo Plus Activator + Nexus 2 (ACT + NX; Kerr) and Ket-etching gel + Panavia F (KE + P; Kuraray). The experimental groups were classified according to the adhesive approach in self-adhesive (UN), etch-and-rinse (ACT + NX, NX, KE + P, SE + UN and VL when bonded to enamel) and self-etch adhesive luting agents (LM, PLP + UN, PN and VL when bonded to dentin). The specimens were stored for 24h in distilled water at 37°C prior to μTBS testing. The Kruskal–Wallis test was used to determine pairwise statistical differences (p < 0.05) in μTBS between the experimental groups.

Results: When bonded to enamel, ACT + NX (15 MPa) and UN (19.6 MPa) scored significantly lower than VL (49.3 MPa), LM (49.2 MPa), PN (35.4 MPa) and SE + UN (35.2 MPa), while PLP + UN (23.5 MPa) showed a significantly lower μTBS than VL (49.3 MPa) and LM (49.2 MPa). No significant differences were noticed between VL (49.3 MPa), LM (49.2 MPa), PN (37.9 MPa), KE + PN (38.8 MPa), PN (35.4 MPa) and SE + UN (35.2 MPa). Regarding the bonding effectiveness to dentin, all luting agents bonded equally effectively (UN: 15.9 MPa; LM: 15.4 MPa; PN: 17.5 MPa; NX: 22.3 MPa; VL: 22.3 MPa), except VL (11.1 MPa) and SE + UN (5.9 MPa) and ACT + NX (13.3 MPa). VL revealed an exceptionally high number of pre-testing failures, most likely due to a combined effect of not having cured the adhesive separately and an insufficiently light-cured luting agent.

Significance: Following a correct application procedure, the etch-and-rinse, self-etch and self-adhesive luting agents are equally effective in bonding to enamel and dentin. Several factors...
1. Introduction

Indirect adhesive procedures constitute a substantial portion of contemporary aesthetic restorative treatments. Tooth-colored inlays, onlays, veneers and crowns are now routinely bonded to the tooth substrate via the use of adhesive resin cements [1–3]. Adhesive resin cements have the ability to bond to both tooth structure and restoration. The integration produces reinforcement of both structures [4, 5], and reduces microleakage at the restoration-tooth interface, postoperative sensitivity, marginal staining and recurrent caries.

Tooth-colored restorations can be constructed in a relatively simple way by CAD-CAM techniques using prefabricated ceramic or composite blocks manufactured under controlled conditions. Indirect composite restorations are now more routinely used because composite blocks show some advantages over ceramic blocks as these have easier finishing and polishing, kinder to the natural dentition with regard to wear and easier to make add-on adjustment. In addition, higher bond strengths to pre-treated composite compared with pre-treated ceramic are reported in vitro [6], probably due to a more compliant situation as the modulus of elasticity of the luting agent and the composite are in the same range. Consequently, several in vitro studies found a more uniform stress distribution throughout teeth restored with indirect composite inlays, compared to ceramic inlays [7, 8].

Besides the advantages of the indirect composite restoration, bonding to tooth structure is still a challenging matter, as the indirect restorative procedure will double the adhesive interfaces. One interface is at the tooth structure and the other at the fitting surface of the restoration. In order to establish a strong and durable bond, which is necessary for the biomechanical aspect of the tooth-restoration system, appropriate treatment of the respective surfaces is crucial.

Adhesion of resin cement to processed composites has traditionally been difficult to achieve [9]. Roughening the composite surface by bur or by sandblasting, followed by silanization has been recommended as a predictable means for enhancing retention between resin cements and the indirect composite restoration [6, 10].

At the tooth surface, an adhesive system is used to bond the luting agent to the tooth substrate. Currently, all adhesives are categorized as either etch-and-rinse or self-etch adhesives [11]. A multi-step application technique is time consuming and rather technique sensitive, and consequently may compromise bonding effectiveness [12]. Recently, a self-adhesive universal resin cement without surface pre-treatment has been introduced (RelyX Unicem; 3 M ESPE, Seefeld, Germany). This self-adhesive resin cement is based on a new monomer, filler and initiation technology. The manufacturer purports that the organic matrix consists of newly developed multifunctional phosphoric acid methacrylates. The phosphoric acidic methacrylates can react with the basic fillers in the luting cement and the hydroxyapatite of the hard tooth tissue.

Several in vitro studies reported the bond strength of different adhesive systems used in combination with a luting composite to both enamel and dentin [13–17]. Little information, however, is available in the literature with regard to the bond strength of the complete tooth/indirect restoration complex using different luting composites categorized by their adhesive system. The purpose of this study was to assess the bonding effectiveness of five adhesive luting agents to enamel and dentin with different application procedures using a standard micro-tensile bond strength (μTBS) test set-up. The hypothesis tested in this study was that there were no differences in bonding effectiveness between etch-and-rinse, self-etch and self-adhesive luting agents.

2. Materials and methods

Forty-two non-carious human third molars (gathered following informed consent, approved by the Commission for Medical Ethics of the Catholic University of Leuven) were stored in 0.5% chloramine in water at 4 °C and used within 1 month after extraction. The teeth were randomly divided into 17 experimental groups.

Specimen preparation is schematically shown in Fig. 1. Table 1 shows the list of adhesive luting agents with their respective application procedures investigated. The general composition of the adhesive luting agents is described in Table 2.

3. Enamel specimen preparation for μTBS

Lingual and/or burcal enamel was flattened using a high-speed diamond bur (642, Komet, Lemgo, Germany), mounted in a Microspecimen Former (University of Iowa, Iowa City, IA, USA). Prior to cementing procedures CAD/CAM composite blocks (max. 7 mm thick; Paradigm MZ100, 3 M ESPE, St. Paul, MN, USA) were cut using a low-speed diamond saw (Isomet 1000, Buehler, Lake Bluff, IL, USA) to obtain a standard surface roughness. The bonding surface was prepared according to the manufacturer’s instructions for the respective cement.

For some luting agents an additional experimental group was made by modifying the adhesive bonding procedure. In total, nine experimental groups were made, classified according to the category of adhesive in etch-and-rinse, self-etch and self-adhesive luting agents (Table 1).

Next, the resin cement was applied on the enamel surface after surface treatment according to the manufacturer’s instructions. The CAD/CAM composite block was pressed on the cement using light pressure, after which excess cement was removed. Light-curing was performed from four parallel
4. Dentin specimen preparation for μTBS

The occlusal third of the molars was removed using the Isomet slow-speed diamond saw. Dentin surfaces were controlled for absence of enamel and/or pulp tissue using a stereomicroscope (Wild MSA, Heerbrugg, Switzerland). The teeth were mounted in a chuck and a standard smear layer was produced by removing a thin layer of the surface using a high-speed medium grit (100 µm) diamond bur (842, Komet) mounted in the MicroSpecimen Former. The cements were subsequently applied following the methodology described by the manufacturer. Similar to the enamel specimens, a modified application procedure was added for some luting agents. In total, eight experimental groups were prepared for testing (Table 1).

5. μTBS testing

After 24h of water storage, the teeth were sectioned perpendicular to the adhesive-tooth interface using the Isomet saw to obtain rectangular sticks of about 2 mm × 2 mm width and 8–9 mm long. Specimens were trimmed at the biomaterial-tooth surface to a cylindrical hourglass shape with a diameter of about 1.2 mm using the MicroSpecimen Former and fine cylindrical diamond burs (Ø = 1.2 mm, RSKRFK, Komet, Lemgo, Germany) under continuous air/water spray. Next, the cross-sectional diameter was precisely measured (accuracy = 0.001 mm) using a precision measuring instrument transformed from an x–y multi-purpose modular microscope (Leitz, Wetzlar, Germany). Specimens were then fixed to Ciucchi’s jig (fixation height = 4–6 mm) with cyano-acrylate glue (Model Repair II Blue, Dentsply-Sankin, Ohtawara, Japan) and stressed at a crosshead speed of 1 mm/min until failure in a LRX material testing device (LRX, Lloyd, Hampshire, UK). The μTBS was expressed in MPa, as derived from dividing the imposed surface (N) at the time of fracture by the bond area (mm²). When specimens failed before actual testing, the μTBS was determined from the specimens that survived specimen processing with an explicit note of the number of pre-testing failures. For the dentin and the enamel side, Kruskal–Wallis test was used to determine pairwise statistical differences (p < 0.05) in μTBS between the experimental groups. All statistics were performed using the Statistica software package (StatSoft, OK, USA).

The mode of failure was determined at a magnification of 50× using a stereomicroscope (Wild MSA).

6. Results

The mean μTBS values are graphically presented per experimental group in box-whisker plots in Fig. 2 (enamel) and Fig. 3 (dentin). Statistically significant differences are mentioned in Table 3. Figs. 4 and 5 show a graphical presentation of proportional prevalence of failure modes for all experimental groups when bonded to enamel (Fig. 4) and dentin (Fig. 5).
Table 1 – List of luting agents and their respective adhesive application procedures

<table>
<thead>
<tr>
<th>Code</th>
<th>Product name</th>
<th>Manufacturer</th>
<th>Enamel/dentin pre-treatment</th>
<th>Composite pre-treatment</th>
<th>Luting agent mixing</th>
</tr>
</thead>
</table>
| Self-adhesive  
UN | RelyX Unicem | 3M ESPE, Seefeld, Germany | No pre-treatment | No pre-treatment | Mix the capsule for 15 min (Rotomix, 3M ESPE), apply on surface, lute resin block, light cure for 20 min from each side |
| Self-etch  
LM | Linkmax | GC, Tokyo, Japan | Mix one drop of each primer liquids EP-A and EP-B for 5 s, apply, air dry after 30 s | Apply GC Etchant liquid for 30 s, rinse, air dry, mix one drop of each Ceramic Primers A and B for 5 s, apply, air dry | Mix A-paste and B-paste for 10–20 s, light cure for 20 s from each side |
| PLP + UN | Prompt L-Pop + RelyX Unicem | 3M ESPE, Seefeld, Germany | Apply Prompt L-Pop, rubbing for 15 s, air blow to thin film, light cure for 10 s | No pre-treatment | Mix the capsule for 15 s (Rotomix, 3M ESPE), apply on surface, lute resin block, light cure for 20 s from each side |
| PN | Panavia-F | Kuraray, Osaka, Japan | Mix one drop of each ED Primer liquids A and B for 5 s, apply, air dry gently after 60 s | Apply K-Etchant gel for 5 s, rinse, air dry, mix one drop of each Cleafil SE Primer and Porcelain Bond Activator for 5 s, apply | Mix universal and catalyst paste for 20 s, light cure for 20 s from each side after removal excess cement, apply oxyguard for 3 min |
| VL | Variolink II | Ivoclar-Vivadent, Schaan, Liechtenstein | Apply Total Etch (37% phosphoric acid) only on enamel for 15 s, rinse, air dry, apply Syntac primer for 15 s, air dry, apply Syntac adhesive for 30 s, air dry, apply HelioBond | Apply Monobond-S for 60 s, air dry, apply HelioBond | Mix base and catalyst paste, light cure for 20 s from each side |
| Etch-and-rinse  
ACT + NX | Optibond Solo Plus Activator + Nexus 2 | Kerr, Orange, CA, USA | Apply Kerr Gel Etchant (35% phosphoric acid) for 15 s, rinse, air dry, mix one drop of Optibond Solo Plus and Optibond Solo Plus Activator for 3 s, apply for 15 s, air dry | Apply Kerr Gel Etchant for 15 s, rinse, air dry, apply Silane Primer, air dry | Mix base and catalyst paste for 10–20 s, light cure for 40 s from each side |
| NX | Nexus 2 | Kerr, Orange, CA, USA | Apply Kerr Gel Etchant (37% phosphoric acid) for 15 s, rinse, air dry, apply Optibond Solo Plus for 15 s, air dry, light cure for 20 s | Apply Kerr Gel Etchant for 15 s, rinse, air dry, apply Silane Primer, air dry | Mix base and catalyst paste for 10–20 s, light cure for 40 s from each side |
| KE + PN | K-Etchant gel + Panavia-F | Kuraray, Osaka, Japan | Apply K-Etchant gel (60% phosphoric acid) on enamel for 10 s, rinse, air dry, mix one drop of each ED Primer liquids A and B for 5 s, apply, air dry gently after 60 s | Apply K-Etchant gel for 5 s, rinse, air dry, mix one drop of each Cleafil SE Primer and Porcelain Bond Activator for 5 s, apply, air blow, polymerization for 10 s | Mix universal and catalyst paste for 20 s, light cure for 20 s from each side after removal excess cement, apply oxyguard for 3 min |
| SE + UN | Scotchbond Etchant + RelyX Unicem | 3M ESPE, Seefeld, Germany | Apply Scotchbond Etchant (35% phosphoric acid) for 15 s, rinse, air dry | No pre-treatment | Mix the capsule for 15 s (Rotomix, 3M ESPE), apply on surface, lute resin block, light cure for 20 s from each side |

* VL: self-etch adhesive that requires selective etching of enamel with phosphoric acid.

* Condition ‘KE + PN’ was not applied for dentin.
Table 2 – General composition of adhesive luting agents tested in this study

<table>
<thead>
<tr>
<th>Material (manufacturer)</th>
<th>Composition</th>
<th>Batch no.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Linkmax (GC Corp., Tokyo, Japan)</td>
<td>Primer A: 4-META, HEMA, dimethacrylate, water, ethanol</td>
<td>301101</td>
</tr>
<tr>
<td></td>
<td>Paste A: fluoroaluminosilicate glass, urethane dimethacrylate, silica, photo-initiator</td>
<td>301091</td>
</tr>
<tr>
<td></td>
<td>Primer B: ethanol, initiator</td>
<td>301101</td>
</tr>
<tr>
<td></td>
<td>Paste B: fluoroaluminosilicate glass, urethane dimethacrylate, silica, photo-initiator</td>
<td>301091</td>
</tr>
<tr>
<td>Nexus 2 (Kerr Co., Orange, CA, USA)</td>
<td>Primer A: ethanol, Bis-GMA, barium aluminoborosilicate glass, silica, sodium hexafluorosilicate</td>
<td>203D20</td>
</tr>
<tr>
<td></td>
<td>Paste A: fluoroaluminosilicate glass, urethane dimethacrylate, silica, photo-initiator</td>
<td>204A61</td>
</tr>
<tr>
<td></td>
<td>Paste B: fluoroaluminosilicate glass</td>
<td>112787</td>
</tr>
<tr>
<td>Panama I (Kuraray Medical Inc., Tokyo, Japan)</td>
<td>Primer A: HEMA, 10-MDE, 5-NMSA, water, accelerator</td>
<td>00108B</td>
</tr>
<tr>
<td></td>
<td>Primer B: 5-NMSA, water, sodium benzene</td>
<td>00115B</td>
</tr>
<tr>
<td></td>
<td>Paste A: 10-MDE, 5-NMSA, silica, dimethacrylate monomer, photo-initiator, accelerator</td>
<td>00124A</td>
</tr>
<tr>
<td></td>
<td>Paste B: barium glass, sodium fluoride, dimethacrylate monomer, RPO</td>
<td>00046A</td>
</tr>
<tr>
<td>RelyX Unicem (3M ESPE, Seefeld, Germany)</td>
<td>Powder: glass powder, silica, calcium hydroxide, pigment, substituted pyrimidine, peroxy compound, initiator</td>
<td>001/0001</td>
</tr>
<tr>
<td></td>
<td>Liquid: methacrylated phosphoric ester, dimethacrylate, acetate, stabilizer, initiator</td>
<td>001/0001</td>
</tr>
<tr>
<td></td>
<td>Scotchbond Etchant: 35% phosphoric acid, silica thickener</td>
<td>001/0001</td>
</tr>
<tr>
<td>Prompt L-Pop (3M ESPE, Seefeld, Germany)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Variolink II (Ivoclar-Vivadent, Schaan, Liechtenstein)</td>
<td>Syntac primer: maleic acid, TEGDMA, water, acetone</td>
<td>D58446</td>
</tr>
<tr>
<td></td>
<td>Syntac adhesive: maleic acid, TEGDMA, glutaraldehyde, water</td>
<td>D59471</td>
</tr>
<tr>
<td></td>
<td>Heliobond: Bis-GMA, TEGDMA</td>
<td>D61246</td>
</tr>
<tr>
<td></td>
<td>Paste A: Bis-GMA, urethane dimethacrylate, TEGDMA, inorganic filler, ytterbium trifluoride, initiator, stabilizer</td>
<td>D61049</td>
</tr>
<tr>
<td></td>
<td>Paste B: Bis-GMA, urethane dimethacrylate, TEGDMA, inorganic filler, ytterbium trifluoride, benzoyl peroxide, stabilizer</td>
<td>D65049</td>
</tr>
</tbody>
</table>

At the enamel, the lowest µTBS values were measured for ACT + NX (15 MPa), UN (19.6 MPa) and PLP + UN (23.5 MPa) with no significant differences between these three groups. The highest µTBS values were obtained with VL (49.3 MPa) and LM (49.2 MPa), while NX (37.9 MPa), PN (35.4 MPa), KE + PN (38.8 MPa) and SE + UN (35.2 MPa) scored in between. No statistically significant differences were noted between these latter six experimental groups. ACT + NX and UN scored significantly lower than LM, PN, SE + UN and VL, while for PLP + UN the difference was only significant with LM and VL.

Failure analysis of the µTBS fracture surfaces demonstrated that when bonded to enamel most UN specimens

Table 3 – P-values from pairwise statistical comparisons (Kruskal–Wallis test)

<table>
<thead>
<tr>
<th>Dentin</th>
<th>Enamel</th>
<th>UN</th>
<th>ACT + NX</th>
<th>KE + PN</th>
<th>SE + UN</th>
<th>VL</th>
<th>LM</th>
<th>PN</th>
<th>PLP + UN</th>
</tr>
</thead>
<tbody>
<tr>
<td>UN</td>
<td>0.819</td>
<td>0.114</td>
<td>0.077</td>
<td>0.934</td>
<td>0.189</td>
<td>0.026</td>
<td>0.026</td>
<td>0.005</td>
<td>0.056</td>
</tr>
<tr>
<td>NX</td>
<td>0.7402</td>
<td>0.942</td>
<td>&gt;0.999</td>
<td>&gt;0.999</td>
<td>&gt;0.999</td>
<td>0.7044</td>
<td>0.7062</td>
<td>0.997</td>
<td>0.3396</td>
</tr>
<tr>
<td>ACT + NX</td>
<td>0.9118</td>
<td>0.2109</td>
<td>---</td>
<td>0.0123</td>
<td>0.0123</td>
<td>0.0123</td>
<td>0.0123</td>
<td>0.0123</td>
<td>0.0123</td>
</tr>
<tr>
<td>KE + PN</td>
<td>---</td>
<td>---</td>
<td>0.077</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>SE + UN</td>
<td>0.0011</td>
<td>0.0053</td>
<td>0.029</td>
<td>0.078</td>
<td>0.1133</td>
<td>0.1133</td>
<td>0.9999</td>
<td>0.0574</td>
<td>0.425</td>
</tr>
<tr>
<td>VL</td>
<td>0.003</td>
<td>0.006</td>
<td>0.029</td>
<td>0.078</td>
<td>0.1133</td>
<td>0.1133</td>
<td>0.9999</td>
<td>0.0574</td>
<td>0.425</td>
</tr>
<tr>
<td>LM</td>
<td>&gt;0.999</td>
<td>0.8916</td>
<td>0.8326</td>
<td>---</td>
<td>0.0408</td>
<td>0.0408</td>
<td>0.0408</td>
<td>0.0408</td>
<td>0.0408</td>
</tr>
<tr>
<td>PN</td>
<td>0.9984</td>
<td>0.9555</td>
<td>0.5176</td>
<td>---</td>
<td>0.0001</td>
<td>0.0269</td>
<td>0.0269</td>
<td>0.0001</td>
<td>0.0269</td>
</tr>
<tr>
<td>PLP + UN</td>
<td>0.5962</td>
<td>0.9997</td>
<td>0.0017</td>
<td>---</td>
<td>0.0001</td>
<td>0.0001</td>
<td>0.0001</td>
<td>0.0001</td>
<td>0.0001</td>
</tr>
</tbody>
</table>

In the right upper half of the table (above asterisks), p-values of comparisons between enamel µTBS are presented. In the left lower half of the table (below asterisks), p-values of comparisons between dentin µTBS are presented. p-values undecorated with an uninterrupted line are smaller than 0.05 and thus indicate significant difference. p-values with an interrupted line represent nearly statistically significant difference.
Fig. 3 – Box plot of dentin μTBS results. The box represents the spreading of the data between the first and third quartile. The central vertical line represents the median. The whiskers extend to the minimum and maximum value measured. ptf/n = number of pre-testing failures/total specimen number.

(78.6%) showed adhesive failures at the luting-enamel interface. For LM, cohesive failures in enamel were the predominant type of failure (78.6%), while the ACT + NX specimens failed mainly (70%) cohesively in the luting agent. For the other luting agents (NX, PN, VL, KE + PN and SE + UN), the failure pattern was a mixture of several types of failure.

Regarding the bonding effectiveness to dentin, all luting agents bonded equally effectively (LM: 15.4 MPa; NX: 22.3 MPa; PN: 17.5 MPa; UN: 15.9 MPa), except VL (1.1 MPa), SE + UN (5.9 MPa) and ACT + NX (13.2 MPa). VL revealed an exceptionally high pre-testing failure number (10/12). The highest bond strengths were measured for NX (22.3 MPa), PLP + UN (22.2 MPa), while LM (15.4 MPa), PN (17.5 MPa) and UN (15.9 MPa) scored in between. Among these latter five groups, no significant differences were noticed, except for PLP + UN, which scored significantly higher than UN.

Concerning the failure modes at the dentin, LM (90%), PN (80%) and SE + UN (100%) specimens failed predominantly adhesively between dentin substrate and luting agent, while in the NX group, most specimens failed (81.8%) adhesively at the luting-Paradigm interface. Next, PLP + UN and ACT + NX showed a 100% and 70% (respectively) cohesive failure in the luting agent. Finally, application of UN and VL to dentin resulted in a mixture of failures at the luting-dentin interface.

7. Discussion

Indirect composite restorations are preferred to direct composite restorations when restoring large tooth defects because of several advantages such as better marginal adaptation and anatomic form, more resistance to wear, increased frac-
The longevity of the indirect composite restoration is influenced by the physico-mechanical properties of the restoration and luting agent, but mainly by the bonding effectiveness at the tooth/restoration complex [21]. The purpose of this in vitro study was to evaluate the bonding effectiveness of this complex using five luting composites in combination with different application procedures.

The luting cements tested in this study were all dual-cure resin cements. Their polymerization is initiated by light and chemically and are therefore the materials of choice to lute indirect tooth-colored restorations with a thickness >3 mm [22,23]. When these dual-cure resin cements are light polymerized, the highest conversion rate is reached [23], with a consequent increase in the physico-mechanical properties [24–26]. Paradigm CAD/CAM composite blocks with a thickness of 7 mm were used in this study as a standard indirect composite restoration. The kind of surface treatment is important to strongly bond to such CAD/CAM generated composite material [27]. The Paradigm surface in the present study was treated according to the instructions of the respective manufacturer of each luting composite tested. For most luting agents (NX, PN and LM), a standard surface roughness was produced by cutting the blocks with a low speed diamond saw, followed by etching with phosphoric acid and silanization, as it is also recommended by several authors [6,10]. The most essential contribution to this bond is suggested to result from the silane coupling agent [17,28]. Using Variolink II (Ivoclar-Vivadent, Schaan, Liechtenstein), the composite surface was silanized, followed by application and polymerization of an adhesive resin. The limited micro-mechanical retention might be responsible for the relatively low bond strength to enamel measured in this study. Indeed, acid etching of enamel with phosphoric acid prior to application of the luting cement significantly increases the enamel bond strength [29].

Regarding the bonding effectiveness to enamel, the lowest scores were measured for the three experimental groups ACT+NX, SE+UN, KE+PN and VL when bonded to enamel, self-etch (LM, PN, PLP+UN and VL when bonded to dentin) and self-adhesive (UN) luting agents.

The limited micro-mechanical retention might be responsible for the relatively low bond strength to enamel measured in this study. Indeed, acid etching of enamel with phosphoric acid prior to application of the luting cement significantly increased the enamel bond strength. Also pre-treatment of the enamel with a strong one-step self-etch adhesive, Prompt L-Pop (3 M ESPE, Seefeld, Germany), increased the bond strength slightly but not significantly. Although TEM and SEM photomicrographs of an enamel surface treated with Prompt L-Pop showed an etching pattern resembling that of the etch-and-rinse adhesives [33–35], several in vitro studies reported somewhat lower bond strength of Prompt L-Pop to enamel as compared to that of etch-and-rinse adhesives [33–36]. These results are in accordance with the results of the present study.

The highest bond strengths to enamel were obtained with PN, LM, SE+UN, KE+PN, NX and VL with no significant differences amongst these six groups. So, the results of this study indicate that the etch-and-rinse (NX, KE+PN, SE+UN and VL) and self-etch approaches (LM and PN) are equally effective in bonding to enamel (except for the two experimental groups mentioned above: ACT+NX and UN). The self-etch adhesive luting cements LM and PN are mild one-step self-etch adhesives, producing a much less micro-retentive etching pattern on enamel as compared to that produced by etch-and-rinse adhesives [37–40]. Nevertheless, several laboratory studies demonstrated relatively high initial bond strengths to instrumented enamel using mild self-etch adhesives [33,41–43]. Some authors found a lower bonding effectiveness when compared to etch-and-rinse adhesives [39,41,43,44], while others reported a similar bonding effectiveness [40,42,45,46] as in the present study. This was clearly demonstrated for Panavia-F (Kuraray, Osaka, Japan), showing similar bond strengths with (KE+PN) and without (PN) primer enamel phosphoric acid etching. Both experimental groups (PN and KE+PN) showed a mixture of several types of failure. Using Linkmax (GC, Tokyo, Japan) (LM), most of the failures after micro-tensile testing were cohesive in enamel, confirming the relatively strong bond of LM to enamel. One has to take into account that the bond strength values measured in this study are the values after 24 h of water stor-
age. An in vitro study of Frankenberger and Tay [47] reported a significant decrease in gap-free margins at the enamel margin of Class-II composite restorations placed using a one-step self-etch adhesive after thermo-mechanical fatigue loading. This was probably attributed to the fact that all-in-one adhesives are more susceptible to water sorption and behave as permeable membranes after polymerization [48,49]. Future studies need to find out how well one-step self-etch adhesive luting agents resist durability testing.

At the dentin, the lowest bond strength was measured for VL with 10 out of 12 specimens having failed prior to testing. The first reason for this low bond strength is that the adhesive was not separately light-cured before application of the luting cement. Indeed, additional light-curing of the adhesive Syntac and HelioBond (Vivacor-Vivadent, Schaan, Liechtenstein) prior to cementation has been demonstrated to increase the dentin bond strength [50] as well as to improve the marginal adaptation to dentin or to the restoration [51]. However, separately light-curing the bonding agent may increase resin thickness and thus affect accurate fit of the restoration in the prepared cavity [50]. That might be the reason why Variolink/Syntac is recommended to lute inlay or crown restorations without prior polymerization of the bonding layer.

Several authors recommended a dual-bonding technique as an alternative to light-curing the adhesive before application of the luting agent without interfering with the fit of the restoration [14,52,53]. This technique involves the application and polymerization of the adhesive to freshly prepared dentin immediately after cavity preparation before taking the impression. It was shown to considerably increase the bond strength values. However, no long-term clinical data for tooth-colored inlays bonded with this technique are available at this moment.

A second reason for the unfavourable low bond strength of VL to dentin is the 7 mm thickness of the Paradigm blocks. The light intensity is drastically reduced when light is transmitted through a 7 mm thick composite block, so that the luting cement must rely almost completely on its self-cure capability. In vitro studies reported a lower auto-polymerizable potential for Variolink II, as compared with several other dual-cure cements [23-25]. This leads to an insufficient conversion rate when used in a self-cure mode. Indeed, Variolink II appears to require enough light to obtain an adequate polymerization efficiency rate and bond strength, as was confirmed by additional testing using 3 mm thick Paradigm composite blocks (unpublished data).

Acid etching prior to application of RelyX Unicem was detrimental for effective dentin bonding. The bond strength of RelyX Unicem to phosphoric acid-etched dentin was significantly lower than for PN, LM, NX, PLP + UN and VL. This must be attributed to inadequate infiltration of the thick and compact collagen mesh (exposed by phosphoric acid) by the viscous cement, as was previously revealed by Fe-SEM and TEM [32]. A weak layer of hydroxyapatite-depleted collagen remained in between RelyX Unicem and the unaffected dentin, and explains the 100% adhesive failure rate in this experimental group. Application of RelyX Unicem without pre-treatment (as recommended by the manufacturer), however, gives a significantly higher bond strength to dentin. The bonding mechanism of this cement is not yet completely understood, but differs from that obtained with self-etch adhesives as no distinct demineralization and hybridization was observed during TEM morphological interface examination [32,54].

Pre-treatment of dentin with the strong one-step self-etch adhesive Prompt L-Pop further significantly increases the bond strength to dentin (PLP + UN). The bond strength was not significantly different from that of LM, PN and NX. On the contrary, many laboratory [36,55,56] and (short-term) clinical studies [57-59] noticed an inefficient bonding effectiveness for this particular adhesive. Several explanations such as incomplete wetting and an insufficiently thick adhesive layer, phase separation between hydrophilic and hydrophobic ingredients, resultant sensitivity to hydrolysis and inhibition of polymerization of the restorative composite on top due to the high acidity of Prompt L-Pop at the oxygen-inhibited layer, have been advanced to explain this inconsistent bonding performance of Prompt L-Pop to dentin. The additional application of a luting composite (on top of Prompt L-Pop) may have served as a conventional hydrophobic resin layer removing several of the above-mentioned explanations for the relatively low bonding effectiveness of the sole use of Prompt L-Pop, thereby explaining the more favourable bonding effectiveness registered in this study. Nevertheless, the 100% cohesive failure rate in the luting cement might still be due to incomplete polymerization of the luting cement, possibly affected by the high acidity of Prompt L-Pop.

The highest bond strength to dentin was obtained with the light-curing adhesive Optibond Solo Plus in combination with the primer with Nexus 2 (NX). Most failures (81.8%) in this group were adhesive at the luting-Paradigm interface, indicating a high bond strength of the luting cement to dentin. No significant differences in bond strengths to dentin were noticed between this etch-and-rinse luting composite (NX), the self-etch luting agents (LM, PN and PLP + UN) and the self-adhesive cement (UN). Nevertheless, their failure modes were different. All specimens using the self-etch adhesive luting agents PN and LM showed an adhesive failure at the luting-dentin interface. This failure mode was also observed in several other in vitro investigations [12,13,16,60] and was explained by the fact that one-step self-etch adhesives, because of their higher concentration of hydrophilic monomers and the lack of the subsequent application of a more hydrophobic resin coating, behave as semi-permeable membranes after polymerization [61]. In addition, as relatively less hydrophilic monomers are available on the tooth surface after application of a one-step self-etch adhesive, the mechanical strength of the adhesive decreases and impairs the bond strength [35,62]. Covering the primed dentin with a more hydrophobic adhesive layer, that is separately light-cured before cementation, increases the bond strength [13,14,16,63-65] and decreases the presence of nanoleakage within the hybrid layer [13]. We may conclude that the results of the study support the tested hypothesis that the etch-and-rinse, self-etch and self-adhesive luting agents showed an equal bond strength to both enamel and dentin, but only on the condition that a correct adhesive procedure was carried out.

Finally, discussing the clinical implications of the results of this study, one must bear in mind that the testing procedure is more simplified than an in vivo situation. The composite
block was bonded to a flat surface, which represents a situation of maximum compliance as there is unrestricted shrinkage strain (i.e. free curing contraction) of the resin cement. In clinical circumstances, when placing an indirect inlay, onlay or crown, the configuration factor will be higher, which may impact the bond strength of the luting cement [37,66]. Therefore, further studies are needed to determine the effect of polymerization contraction stress on the bonding effectiveness of indirect restorations in prepared cavities. In addition, as the durability of the bond greatly determines the longevity of the restoration, there is certainly a need to test the bond durability of adhesive luting agents in vitro and in vivo.

8. Conclusions
An adequate bonding effectiveness to tooth structure can be obtained after 24 h of water storage with all luting agents tested, on the condition that a correct adhesive procedure was carried out. Some factors negatively influenced the bonding effectiveness; these are: (1) not separately light-curing the adhesive before luting; (2) use of a light-cure adhesive converted into a dual-cure version; (3) use of a dual-cure luting agent with low auto-polymerizable potential; (4) acid etching dentin with phosphoric acid prior to the use of RelyX Unicem; (5) bonding of RelyX Unicem to enamel without prior phosphoric acid etching. Taking these factors into account, the etch-and-rinse, self-etch and self-adhesive luting cements tested in this study are equally effective to bond to enamel and dentin, at least in the short term.

**REFERENCES**


