Effect of low-shrinking composite on the bonding effectiveness of two adhesives in occlusal Class-I cavities

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The purpose of this study was to evaluate if a low-shrinking composite can improve the bonding effectiveness of adhesives in highly constrained conditions. A low-shrinking composite (‘els-extra low shrinkage’, Saremco) was bonded in standardized occlusal Class-I cavities using a three-step (‘cmf’, Saremco) and a two-step etch-and-rinse adhesive (‘XP Bond’, Dentsply). Both adhesives were also combined with a conventional composite (‘Z100’, 3M ESPE). Half of the restored cavities were exposed to 20,000 thermo-cycles. 3-way ANOVA revealed a significant effect for the factors ‘adhesive’ and ‘composite’ (both $p<0.0001$), but not for ‘thermo-cycling’ ($p=0.994$). Significantly higher bond strengths were recorded for the low-shrinking composite than for the control composite, using either of the adhesives. The low-shrinking composite in combination with the three-step etch-and-rinse adhesive performed best in the high C-factor Class-I cavity. The two-step etch-and-rinse adhesive suffered strongly from polymerization-shrinkage stress, which could be partially restored by using the low-shrinking composite.

Keywords: Class-I cavity, Low-shrinking composite, Bond strength, Etch and rinse, Thermo-cycling

INTRODUCTION

Since long, polymerization shrinkage and the resulting shrinkage stress are considered major shortcomings of dental composite to restore teeth. Low-shrinking composites should help to avoid clinical problems that are commonly associated with composite restorations, such as post-operative sensitivity, enamel cracks, rapid discoloration and deterioration of restoration margins, early development of caries recurrence, etc. In this respect, many research reports support the general idea that contraction stress due to polymerization shrinkage of the composite results in early failure of especially the bond to dentin that is considered weakest of the restored tooth complex.

The purpose of this study was to evaluate if the use of a low-shrinking composite can improve the bonding effectiveness of adhesives in highly constrained conditions. The hypotheses tested were therefore (1) that the micro-tensile bond strength to Class-I cavity-bottom dentin was higher for a low-shrinking than for a high-shrinking composite, and (2) that the bond obtained using the low-shrinking composite is also more resistant to extended thermo-cycling.

MATERIALS AND METHODS

Specimen preparation

The experimental set-up is schematically presented in Fig. 1. Forty 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 solution at 4°C and used within 1 month after extraction. First, all teeth were mounted in gypsum blocks in order to ease manipulation. Standard box-type Class-I cavities (4.5×4.5 mm wide, 2.5 mm deep) were prepared at the occlusal crown center with the pulpal floor ending at mid-coronal dentin, using a cylindrical medium-grit (100 µm) diamond bur (842, Komet, Lemgo, Germany) in a water-cooled high-speed contra-angle handpiece mounted in the MicroSpecimen Former (The University of Iowa, Iowa City, IA, USA). All cavity surfaces were carefully verified for absence of enamel and/or potential pulp exposure using a stereomicroscope (Wild M5A, Heerbrugg, Switzerland). The teeth were randomly divided into 4 experimental groups.

The adhesives and the composites were then applied, strictly according to the manufacturer’s instructions (Table 1). A low-shrinking composite (‘els-extra low shrinkage’, Saremco, St. Gallen, Switzerland) was bonded into the standardized occlusal Class-I cavities using either a three-step (‘cmf’, Saremco) or two-step etch-and-rinse adhesive (‘XP Bond’, Dentsply, Konstanz, Germany). Both adhesives were also combined with a conventional composite (‘Z100’, 3M ESPE, Seefeld, Germany) that is known to shrink significantly more during polymerization, and thus served as control. All cavities were filled in bulk, after which the composite was light-cured for 40 s using a high-power LED light-curing device (L.E.Demetron I, Demetron/Kerr, United States).
After bonding procedures, the restored teeth were stored for 1 day in distilled water at 37°C. Next, the specimens were randomly divided into two groups, of which half (5 teeth/group) were subjected to 20,000 thermo-cycles (in water baths of 5 and 55°C with a dwell time of 30 s; Thermocycler, Willytec, Munich, Germany), and the other half were stored for 3 weeks at 37°C. The latter time equals the time needed for the 20,000 thermo-cycles.

Micro-tensile bond strength (μTBS) testing
After artificial aging, the teeth were sectioned perpendicular to the adhesive-tooth interface using an automated precision water-cooled diamond saw (Accutom-50, Struers A/S, Ballerup, Denmark) to obtain rectangular sticks (1.0×1.0 mm wide, 5–8 mm long). All specimens from each group were carefully checked using the stereo-microscope and specimens containing angulated interfaces and/or parts of the cavity wall were excluded. Specimens were then fixed to the BIOMAT μTBS-jig with cyanoacrylate glue (Model Repair II Blue, Dentsply-Sankin, Tochigi, Japan) and stressed at a crosshead speed of 1 mm/min until failure in a testing device (LRX, Lloyd, Hampshire, UK) using a load cell of 100 N. The μTBS was expressed in MPa, as derived from dividing the imposed force (N) at the time of fracture by the bond area (mm²). The actual number of pre-testing failures (ptf) was explicitly noted as well. A two-parameter Weibull regression analysis was conducted to determine the shape and size estimates for every experimental group. These were used to construct probability of failure curves at various stress levels, and in particular at relatively low ones. To explore differences between the different groups in more detail, μTBS data were subjected to a three-way analysis of variance (ANOVA) and post-hoc multi-comparisons tests. All tests were conducted using a statistical software package (Statistica 9.0, StatSoft, Tulsa, OK, USA) at a significance level of 0.05.

Failure analysis
The mode of failure was determined light-microscopically at a magnification of 50× using the stereo-microscope, and recorded as either ‘cohesive failure in dentin’, ‘cohesive failure in resin’, ‘interfacial failure between...
Table 1  Materials used

<table>
<thead>
<tr>
<th>Materials</th>
<th>Composition</th>
<th>Application</th>
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<tbody>
<tr>
<td><strong>Adhesive</strong></td>
<td>cmf Etch [LOT: 11.2011-01]: water, H_2PO_4, phosphoric salt, gel former, colorant; gel buffered to a pH of 1.5.</td>
<td>1) Etch enamel and dentin for 30 s with cmf Etch. 2) Rinse for 30 s and air-dry for 5 s. 3) Apply cmf Primer using a rubbing motion for 30 s, dry for 5 s and light cure for 20 s. 4) Apply cmf Bond using a rubbing motion for 20 s. 5) Light-cure for 30 s.</td>
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<tr>
<td>Saremco, St. Gallen, Switzerland</td>
<td>cmf Primer [LOT: 11.2011-01]: alcohol, acetone, water, methacrylated phosphoric salt, CQ, co-initiator (pH=4.5). cmf Bond [LOT: 11.2011 06]: hydrophilic ethoxylated Bis-GMA, silanized barium glass, CQ, co-initiator.</td>
<td></td>
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<tr>
<td><strong>XP Bond</strong></td>
<td>Dentsply, Konstanz, Germany ExTrey Conditioner 36 [LOT: 0507002143]: 36% H_3PO_4, highly dispersed silicon dioxide, detergent, pigment, water (pH=0.0). Adhesive [LOT: 0702001786]: TCB resin, PENTA, UDMA, TEGDMA, HEMA, butylated benzenedi (stabilizer), ethyl-4-dimethylaminobenzoate, CQ, functionalised amorphous silica, t-butanol.</td>
<td>1) Start etching at enamel margins for minimally 15 s, and extend to dentin for maximally 15 s. 2) Remove acid gel and rinse for minimally 15 s, and remove excess water. Do not desiccate. 3) Wet all surfaces with XP Bond. Avoid pooling. Leave XP Bond undisturbed for 20 s. Evaporate solvent with air for minimally 5 s. 4) Light cure for 20 s (&gt;500 mW/cm^2).</td>
</tr>
<tr>
<td><strong>Composite</strong></td>
<td>els</td>
<td>extra low shrinkage, Shade: A2</td>
</tr>
<tr>
<td>Saremco, St. Gallen, Switzerland</td>
<td>[LOT: 11.2011-52] silanized barium glass, Bis-GMA, Bis-EMA, catalysts, inhibitors, pigments.</td>
<td></td>
</tr>
<tr>
<td>Z100 MP Restorative, Shade: A2</td>
<td>[LOT: 7RR] zirconia/silica filler, Bis-GMA, TEGDMA.</td>
<td>1) The thickness of the individual increments must not exceed 2.5 mm. 2) Light cure for 40 s.</td>
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Table 2  Micro-tensile bond strength in MPa

<table>
<thead>
<tr>
<th>Adhesive</th>
<th>3-week water storage (5 teeth/group)</th>
<th>20,000× thermo-cycling (5 teeth/group)</th>
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<tr>
<td></td>
<td>cmf</td>
<td>XP Bond</td>
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<tr>
<td></td>
<td>els Z100</td>
<td>els Z100</td>
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<tr>
<td>ptf/n</td>
<td>0/20 7/20 3/20 20/20</td>
<td>0/20 1/20 9/20 19/20</td>
</tr>
<tr>
<td>Weibull analysis</td>
<td></td>
<td></td>
</tr>
<tr>
<td>scale</td>
<td>29.19 28.09 18.06 –</td>
<td>–</td>
</tr>
<tr>
<td>descriptive statistics</td>
<td></td>
<td></td>
</tr>
<tr>
<td>ptf=0 MPa</td>
<td>26.2±9.2 16.3±6.9 13.7±12.1 0</td>
<td>27.5±9.2 20.3±13.6 8.3±10.5 0.1±0.4</td>
</tr>
<tr>
<td>ptf excluded</td>
<td>26.2±9.2 25.1±14.6 16.1±11.6 no data</td>
<td>27.5±9.2 21.4±13.1 15.0±9.8 1.7</td>
</tr>
<tr>
<td>ptf=lowest measured</td>
<td>26.2±9.2 17.2±13.0 14.3±11.4 no data</td>
<td>27.5±9.2 20.6±13.3 9.8±9.2 1.7</td>
</tr>
</tbody>
</table>

(mean±SD); ptf=pre-testing failure; n=total number of specimens.
dentin and adhesive’, ‘interfacial failure between bond and composite’, or ‘mixed failure’. Representative samples were processed for field-emission-gun scanning electron microscopy (Feg-SEM; Philips XL30, Eindhoven, Netherlands), using common SEM specimen-preparation procedures\(^{10}\).

**RESULTS**

The mean $\mu$TBS, standard deviation (SD), the number of pre-testing failures (ptf) and the total number of specimens (n) are summarized per group in Table 2, and graphically presented in box-whisker plots in Fig. 2. The Weibull moduli (shape, scale) are also summarized in Table 2, and graphically presented in Fig. 3. The three-step etch-and-rinse adhesive ‘cmf’ (Saremco) in combination with the low-shrinking composite ‘els’ (Saremco) exhibited the highest Weibull modulus, suggesting the best performance at low stress levels and in particular the least technique-sensitive adhesive-composite combination. Interesting to note is the similarity in shape and position of the weibull graphs of the respective control and thermo-cycled groups, indicating that thermo-cycling had no effect in the present study.

Three-way ANOVA analysis revealed a significant effect for the parameters ‘composite’ ($p<0.0001$) and ‘adhesive’ ($p<0.0001$), but not for ‘thermo-cycling’ ($p=0.994$), nor for any correlations between the individual factors(adhesive×composite; $p=0.4700$; adhesive×thermo-cycling: $p=0.1155$; composite×thermo-cycling: $p=1.503$; adhesive×composite×thermo-cycling: $p=0.6786$). During specimen preparation, none of the micro-specimens prepared with the three-step etch-and-rinse adhesive ‘cmf’ (Saremco) in combination with the low-shrinking composite ‘els’ (Saremco) failed before actual testing. When the three-step etch-and-rinse adhesive was combined with either the low- or high-shrinking composite, and when subjected to thermo-cycling or not, no significant differences in bond strength were found. When the two-step etch-and-rinse adhesive ‘XP-Bond’ (Dentsply) was used in combination with the high-shrinking composite ‘Z100’ (3M ESPE), only one single micro-specimen out of 5 teeth survived specimen processing (19 pre-testing failures on a total of 20 specimens), yielding a very low bond strength of 1.7 MPa.

Failure analysis data are graphically presented in Fig. 4. Most fractured surfaces appeared to have failed ‘mixed’ (partially at the interface and partially within the resin), irrespective of the experimental group. Detailed Feg-SEM confirmed this predominantly ‘mixed’ failure mode (Fig. 5). Pre-test failures that could be observed all failed at the resin-dentin interface.

**DISCUSSION**

Hypothesis (1) that the micro-tensile bond strength to Class-I cavity-bottom dentin was higher for the low-shrinking than for the high-shrinking composite was accepted, thereby confirming the general reasoning that high C-factor cavities are better restored with low-shrinking composites. Hypothesis (2) that the bond obtained using the low-shrinking composite is more resistant to thermo-cycling was rejected, as thermo-cycling had no effect at all in this study. Both adhesives did however behave differently; the three-step etch-and-
rinse adhesive ‘cmf’ (Saremco) outperformed the two-step etch-and-rinse adhesive ‘XP-Bond’ (Dentsply), irrespective of the composite they were combined with. This confirms the generally documented better bonding potential of conventional three-step adhesives over their simplified successors\textsuperscript{11,12}.

Artificial aging through thermo-cycling did not appear to decrease the bond strength more than simple water storage for 3 weeks, the time needed for the 20,000 thermo-cycles. Nevertheless, thermo-cycling is a widely used as a laboratory aging methodology following a dedicated ISO standard. The ISO TR 11450 standard\textsuperscript{13} recommends to fatigue specimens by a thermo-cycling regimen of 500 cycles in water between 5 and 55°C. Current literature is however inconclusive on the actual effectiveness of thermo-cycling for aging\textsuperscript{14,15}. This should primarily be attributed to the ISO-recommended 500-cycle regimen that is probably much too weak to actually challenge the bond integrity\textsuperscript{12,16}. A literature review concluded that 10,000 thermo-cycles corresponds approximately to 1 year of in vivo functioning, rendering 500 cycles following the ISO standard as very low\textsuperscript{14}. This is corroborated by a meta-analysis, concerning data published between 1992 and 1996, that concluded that thermo-cycling does not significantly influence bond-strength outcomes\textsuperscript{15}. Most studies included in the meta-analysis were carried out following the ISO standard of 500 cycles (mean number of cycles in the studies applied was 630). However, as appeared from this study, significantly longer thermo-cycling up to 20,000 cycles did also not challenge the specimens more than just water storage did. We thermo-cycled the whole restored teeth; most likely a much longer thermo-cycling protocol should have been applied in order to achieve bond...
degradation. Alternatively, faster artificial aging could have better been achieved when the specimens would have been cut directly into small µTBS-specimens and as such stored in water (and/or exposed to thermocycling).17,18)

In the present study, a µTBS-testing protocol was used to assess adhesion in constrained conditions. Using conventional macro-shear or -tensile tests, such particular bonding effectiveness in Class-I cavities cannot be measured19,20). A known drawback of using micro-specimens is however that they are prone to failure prior to the actual bond-strength test. A current topic of debate thus remains how to correctly interpret pre-testing failures (ptf’s) with regard to the calculation of the average µTBS and further statistical analysis. A first option is to employ a kind of survival analysis, like the Weibull analysis performed in this study; it can deal perfectly with actually missing bond-strength data. For
more conventional analysis, diverse options are available. We have therefore provided a table (Table 2) and graph (Fig. 2), in which (1) a ptf was considered as 0 MPa, which actually penalizes the outcome too severely (as there was a certain bond strength), (2) a ptf was excluded from the average µTBS calculation, and thus part of our data were neglected, and (3) a ptf that received the lowest µTBS measured within the specific experimental group. Depending on how the ptf’s were dealt with, the box plots changed clearly, and the statistical analysis revealed additional information for the specimens that underwent thermo-cycling. Especially when the two-step etch-and-rinse adhesive ‘XP-Bond’ (Denstply) was combined with the low-shrinking composite ‘els’ (Saremco), and the specimens were exposed to thermo-cycling, 9 ptf’s out of 20 specimens were recorded, thereby clearly altering the distribution of the data depending on how the ptf’s were taking into account. In case the ptf’s were excluded, the data were normally distributed, so that an ANOVA analysis could be performed followed by Tukey-Kramer multiple comparisons. In case the ptf’s were considered as 0 MPa or the lowest µTBS measured, the distribution was inevitably skewed so that only non-parametric analyses could be performed. In this particular group, the mean bond-strength value varied considerably from 8.3 MPa, when a ptf was considered as 0 MPa, to 15.0 MPa when all ptf’s were excluded. The post-hoc multiple comparisons were obviously different as well (Fig. 2).

Weibull probability of survival analysis has for instance been developed in engineering as method to ‘design’ ceramic compounds\(^{22}\). One of the great advantages of Weibull analysis is that it allows predicting the likelihood of failure at low stress levels. Theoretically, as such one can design a material following the expected need of resistance against stress it will be exposed to. In case of a restoration in the oral cavity, the needed stress resistance is probably much lower than the measured mean bond-strength values. At 10 MPa for example, the probability of failure for the three-step adhesive ‘cmf’ (Saremco) is only 3% for the low-shrinking composite ‘els’ (Saremco), but 14–20% for the high-shrinking composite ‘Z100’ (3M ESPE). Both failure probabilities differ significantly depending on the shrinkage level of the composite, while the respective mean bond-strength values do not (26.2±9.2 and 25.1±14.6 MPa, respectively). This indicates that the data repeatability and reliability is higher in case a low-shrinkage composite was employed. This also appears from the difference in weibull-shape moduli (3.35 versus 1.79, respectively). Therefore, the adhesive-composite combination with the highest Weibull modulus reflects the best performing and in particular the least technique-sensitive combination\(^{22}\).

Most of the ptf’s occurred when the specimens were cut using the automated precision water-cooled diamond saw in the group that combined the two-step etch-and-rinse adhesive ‘XP-Bond’ (Dentsply) with the high-shrinking composite ‘Z100’ (3M ESPE). Micro-specimens that were excluded for bond-strength measurement because part of the axial cavity wall was included, nevertheless revealed information that may very likely explain the high amount of ptf’s observed (Fig. 6). Such samples indeed showed a clear gap between the bottom of the cavity and the composite, indicating that the stress developed during polymerization of the high-shrinking composite must have been that high that the adhesive was pulled away from dentin in this constrained high C-factor cavity. This very unfavourable bonding contrasts with the relatively high bond strength measured before for the same adhesive/composite combination when bonded to flat dentin surfaces (52.2±10.6 MPa, though without exposure to artificial aging\(^{29}\)), and with data measured elsewhere (38.0±7.8 MPa)\(^ {29}\). The high-shrinking composite ‘Z100’ (3M ESPE), used as control in this study, possesses a high E-modulus (21 GPa)\(^ {29}\). When bonded to a flat dentin surface, the composite can ‘flow’ sufficiently and thus shrink nearly unrestrained without much challenging the adhesive-dentin interface. However, when such a stiff composite is applied in a relatively small Class-I cavity with a high C-factor of 3.2, polymerization shrinkage is hindered much more (less free surface), and therefore must have resulted in a much higher polymerization-shrinkage stress\(^{29}\). It is clear that the two-step etch-and-rinse adhesive ‘XP-Bond’ (Dentsply) appears to withstand such shrinkage stress less good than the three-step etch-and-rinse adhesive ‘cmf’ (Saremco). Besides producing a bond that is less resistant to aging, also other factors like differences in
adhesive-layer thickness and elastic properties may explain the markedly different bonding performance recorded for the two adhesives in this study. Since for instance also the risk on improper drying of etched dentin is much higher at narrow Class-I cavities (than at flat surfaces), adequate solvent removal must also have been more difficult for the combined primer/adhesive resin of the two-step etch-and-rinse adhesive ‘XP-Bond’ (Denstply) than of the separate primer of the three-step system ‘cmf’ (Saremco).

Another marked difference between both adhesives is the pH of the etchant that was respectively applied (Table 1); this definitely resulted in significantly different surface topography and thus also interface ultrastructure27). While the two-step adhesive ‘XP-Bond’ (Denstply) produced a 4-µm thick hybrid layer, the three-step etch-and-rinse adhesive ‘cmf’ (Saremco) on the other hand produced a 1.5–3 µm hybrid layer27). The latter thinner hybrid layer resulted from the use of the buffered (pH=1.5) and thus ‘milder’ phosphoric-acid conditioner (as compared to ‘DeTrey Conditioner 36’, as it was applied with XP-Bond (Dentsply) and contains 36% H3PO4, thereby having a pH near 028). It is currently not clear if this difference in hybrid-layer thickness may also account for the difference in hybridization quality and bond strength measured, although it is generally accepted that bonding effectiveness is not related to hybrid-layer thickness29,30).

It is concluded that the low-shrinking composite ‘els’ (Saremco) in combination with the three-step etch-and-rinse adhesive ‘cmf’ (Saremco) performed best in high C-factor Class-I cavities. The two-step etch-and-rinse adhesive ‘XP-Bond’ (Denstply) suffered strongly from polymerization-shrinkage stress, which could be partially reverted when it was combined with the low-shrinking composite ‘Z100’ (3M ESPE). Interface aging using a 20,000 thermo-cycling regime exposed to the gap formation in Class I restorations. Dent Mater 2004; 20: 694-699.

ACKNOWLEDGMENTS
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REFERENCES


