Universal Adhesives Benefit from an Extra Hydrophobic Adhesive Layer When Light Cured Beforehand


**Purpose:** To measure microtensile bond strength (μTBS) of universal adhesives immediately and after 6-month aging, with or without an additional adhesive layer applied on a separately light-cured or non-light-cured universal adhesive.

**Materials and Methods:** Eighty human third molars were randomly assigned to 8 experimental groups. The universal adhesives Clearfil Universal Bond (Kuraray Noritake) and Single Bond Universal (3M Oral Care) were used in self-etch mode (following the manufacturer’s directions), and either light cured or not before application of an extra hydrophobic adhesive layer (Clearfil SE Bond bond, Kuraray Noritake). The two-step self-etch adhesives Clearfil SE Bond (Kuraray Noritake) and OptiBond XTR (Kerr) were used as references. After composite buildups were bonded to mid-coronal occlusal dentin surfaces, the specimens were stored in water (37°C/24 h) and sectioned into microspecimens (0.96 ± 0.04 mm²). Half of the specimens were immediately subjected to μTBS testing (1.0 mm/min), while the other half was stored in water (37°C) for 6 months prior to testing. Failure analysis was performed using stereomicroscopy and SEM. Data were analyzed with two-way repeated measures ANOVA, Tukey’s and paired t-tests (p = 0.05).

**Results:** The immediate μTBS was similar for the universal adhesives when applied following the different application strategies (p > 0.05). Application of an extra layer of hydrophobic adhesive improved the aged μTBS of the universal adhesives, which was statistically significant when the universal adhesives were first light cured (p < 0.05). The reference adhesives Clearfil SE Bond and OptiBond XTR exhibited a significantly higher immediate and aged μTBS to dentin than did the universal adhesives (p < 0.05).

**Conclusion:** The bond durability of universal adhesives, applied in self-etch mode, was found to benefit from the application of an extra hydrophobic adhesive layer when the universal adhesives were first light cured.

**Keywords:** aging, dentin, microtensile bond strength, self-etch adhesives.

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The trend in current adhesive technology is to simplify bonding procedures by reducing application steps, shortening clinical application time, and decreasing technique sensitivity. Recently, a new generation of adhesives has been introduced. These adhesives are termed “universal” or “multi-mode”, as they can be applied either in an etch-and-rinse or self-etch mode. A third option involves their application on phosphoric-acid pre-etched tooth enamel (and unetched dentin) following the so-called selective enamel-etching approach to achieve better bond durability to enamel, while maintaining the self-etch approach on dentin. The latter adhesive protocol preserves the potential to achieve additional primary chemical bonding with the remaining (carbonated) apatite crystallites in dentin (and enamel). The manufacturers claim that one adhesive solution can be used for either adhesive strategy without compromising bonding efficacy, by which these universal adhesives could replace the previous generation of simplified non-universal adhesives that needed to be applied either following an etch-and-rinse or a self-etch approach. The optional etch-and-rinse or self-etch application mode of universal adhesives has been made possible through the inclusion of hydrophilic functional monomers along with an increased amount of solvent, making these adhesives com-
compatible with both a non-pre-etched (self-etch mode) or pre-etched (etch-and-rinse mode) dentin substrate.\textsuperscript{20,49}

However, the adapted adhesive formulations of universal adhesives have been shown to lead to more residual solvent entrapment within the adhesive layer.\textsuperscript{49} Such residual solvents may hinder the formation of a highly cross-linked polymer, thereby decreasing the polymerization-conversion degree within the adhesive layer, which in turn may affect the adhesive-dentin bond strength and increase the permeability of the adhesive layer after polymerization.\textsuperscript{15,47} Consequently, these effects may compromise the final structure of the polymer and its mechanical properties, making it more vulnerable to accelerated bond degradation.\textsuperscript{15,47,49}
It has been advocated that the short- and long-term bonding efficacy of one-step self-etch adhesives can be improved by the placement of an additional hydrophobic resin coat.\(^1\,13\,24\) Therefore, one method to overcome such deficiencies of universal adhesives involves the application of an additional layer of a hydrophobic resin to coat the adhesive. This extra resin coat increases the thickness and uniformity of the adhesive layer with the expectation to reduce fluid flow across the adhesive interface.\(^1\,13\,24\) However, with a thicker adhesive layer it may be more difficult to volatilize the solvent before light curing and so negatively affect bond strength.\(^56\) The solution to this problem could be to light cure the adhesive layer separately prior to the application of the extra hydrophobic resin. Only a few laboratory studies have reported that placement of a hydrophobic resin coating over the polymerized universal adhesives indeed improved the microtensile dentin bond strength (\(\mu\text{TBS}\)) of universal adhesives when used in self-etch mode.\(^20\,29\) Nevertheless, the effect of applying an additional hydrophobic adhesive layer on a separately light-cured or non-light-cured universal adhesive has not been tested so far.

The aim of this study was therefore to evaluate the immediate (24 h) and aged (6 months) \(\mu\text{TBS}\) of two universal adhesives used in self-etch mode on dentin with or without an additional hydrophobic adhesive layer applied on the separately light-cured or non-light-cured universal adhesive. The hypotheses tested were (1) that the use of an additional hydrophobic adhesive layer applied on a non-light-cured universal adhesive would not improve either the immediate or aged bond strength to dentin, and (2) that the use of an additional hydrophobic adhesive layer applied on a separately light-cured universal adhesive would not improve either the immediate or aged bond strength to dentin.

**MATERIALS AND METHODS**

**Tooth Selection and Preparation**

Eighty extracted caries-free human third molars were used following ethical approval (ref no: 06.01.2016/4). In this study, the teeth were disinfected in 0.5% chloramine, stored in distilled water, and used within three months after extraction. The mid-coronal dentin surface was exposed in all teeth after sectioning the occlusal enamel using a water-cooled diamond saw (Minitom, Struers; Ballerup, Denmark). The dentin surfaces were controlled for absence of enamel and/or pulp tissue using a stereomicroscope (S4E, Leica Microsystems; Wetzlar, Germany). The exposed dentin surfaces were further ground with wet 320-grit silicon carbide paper (MD Fuga, Struers) for 60 s to standardize the preparation of smear layers (Fig 1).

**Experimental Design and Specimen Preparation**

The teeth were randomly assigned to eight groups (n = 10) according to the different adhesive strategies of the respective adhesive (Tables 1 and 2). The two universal adhesives were solely applied in self-etch mode: Clearfil Universal Bond (Kuraray Noritake; Tokyo, Japan) and Single Bond Universal (3M Oral Care; St Paul, MN, USA; also marketed as Scotchbond Universal in other parts of the world).

**Table 1  Adhesives investigated, chemical compositions, and application procedures**

<table>
<thead>
<tr>
<th>Adhesive (manufacturer)</th>
<th>Composition*</th>
<th>Application procedure</th>
</tr>
</thead>
</table>
| Clearfil Universal Bond (Kuraray Noritake; Tokyo, Japan) Lot no: 3D0006 | 10-MDP; bis-GMA, hydrophilic aliphatic DM, HEMA, colloidal silica, camphorquinone, silane, accelerators, initiators, ethanol, water                                                                 | 1. Apply the adhesive to the entire cavity wall with the applicator brush and rub it in for 10 s.  
2. Dry the cavity wall sufficiently by blowing mild air for more than 5 s until the adhesive shows no movement; use a vacuum aspirator to prevent the adhesive from scattering.  
3. Light cure for 10 s.                                                                                                                                              |
| Single Bond Universal** (3M Oral Care; St Paul, MN, USA) Lot no: 609973 | 10-MDP, DM, HEMA, methacrylate-modified polyalkenoic acid copolymer, filler, initiators, silane, ethanol, water                                                                                            | 1. Apply the adhesive to the entire preparation with a microbrush and rub it in for 20 s.  
2. Direct a gentle stream of air over the liquid for about 5 s until it no longer moves and the solvent is evaporated completely.  
3. Light cure for 10 s.                                                                                                                                                        |
| Clearfil SE Bond (Kuraray Noritake) Lot no: 000156 | Primer: 10-MDP hydrophilic DM, HEMA, camphorquinone, water  
Bond: 10-MDP; bis-GMA, hydrophobic DM, HEMA, camphorquinone, N,N-diethanol p-toluidine, colloidal silica | 1. Apply Primer to tooth surface and leave in place for 20 s.  
2. Dry with air stream to evaporate the volatile ingredients.  
3. Apply Bond to the tooth surface and then create a uniform film using a gentle air stream.  
4. Light cure for 10 s.                                                                                                                                                         |
| Optibond XTR (Kerr; Orange, CA, USA) Lot no: 5351340 | Primer: GPDM, hydrophilic co-monomers, ethanol, aceton, water  
Bond: resin monomers, HEMA, inorganic fillers, ethanol | 1. Scrub Primer with a brushing motion for 20 s.  
2. Air thin for 5 s.  
3. Apply Bond for 15 s.  
4. Light cure for 10 s.                                                                                                                                                        |

*Composition as provided by the respective manufacturers. Bis-GMA: bisphenol A-glycidyl methacrylate; DM: dimethacrylate; GPDM: glycerol phosphate dimethacrylate; **marketed as Scotchbond Universal in other parts of the world.
testing jig with cyanoacrylate glue (Zapit, Dental Ventures of America; Corona, CA, USA) and stressed at a crosshead speed of 1 mm/min until failure in a LRX testing device (LR 5K Lloyd Instruments; West Sussex, UK) using a load cell of 100 N. After measuring the exact dimensions of each fractured stick with the digital caliper (Digimatic Caliper), the μTBS was calculated in MPa, as derived from dividing the imposed force (in N) at the time of fracture by the bond area (in mm²). When specimens failed before actual testing (pre-test failures, ptf), they were included as 0 MPa in the calculation of the mean μTBS.

For each experimental group, the mean of μTBSs of the sticks originating from the same tooth were calculated and the mean bond strength was used as one unit for statistical analysis. Data were submitted to two-way repeated measures ANOVA. The adhesive strategy (universal adhesive without a hydrophobic adhesive layer, light-cured or non-light-cured universal adhesive with an extra hydrophobic adhesive layer, two-step self-etch adhesive) and storage time were the two main factors. Tukey’s post-hoc multiple comparison test was used for pairwise comparisons. To compare the means regarding storage time (24 h or 6 months), the paired samples t-test was performed. A significance level of 0.05 was used for all tests.

**Failure Analysis**

The failure mode analysis was performed under a stereomicroscope at 80X magnification. The failure mode was classified as adhesive, mixed, cohesive in composite, cohesive failure in dentin, or as mixed failure when more than one of the fracture modes occurred at the fractured surface. Four representative specimens of each experimental group were selected. Each specimen was fixed onto an aluminum SEM sample holder with carbon glue and observed using scanning electron microscopy (SEM Quanta Feg 250, FEI; Eindhoven, The Netherlands).

### Table 2  Overview of the experimental groups

<table>
<thead>
<tr>
<th>Adhesive</th>
<th>Adhesive strategy</th>
<th>Experimental group code</th>
</tr>
</thead>
<tbody>
<tr>
<td>Universal adhesive used in self-etch mode</td>
<td>Light-cured Clearfil Universal Bond without hydrophobic adhesive layer</td>
<td>CUB</td>
</tr>
<tr>
<td></td>
<td>Non-light-cured Clearfil Universal Bond with light-cured hydrophobic adhesive layer</td>
<td>CUB_nonLC+C-SEbond</td>
</tr>
<tr>
<td></td>
<td>Light-cured Clearfil Universal Bond with light-cured hydrophobic adhesive layer</td>
<td>CUB_LC+C-SEbond</td>
</tr>
<tr>
<td></td>
<td>Light-cured Single Bond Universal without hydrophobic adhesive layer</td>
<td>SBU</td>
</tr>
<tr>
<td></td>
<td>Non-light-cured Single Bond Universal with light-cured hydrophobic adhesive layer</td>
<td>SBU_nonLC+C-SEbond</td>
</tr>
<tr>
<td></td>
<td>Light-cured Single Bond Universal with light-cured hydrophobic adhesive layer</td>
<td>SBU_LC+C-SEbond</td>
</tr>
<tr>
<td>Two-step self-etch adhesive (reference)</td>
<td>Clearfil SE Bond</td>
<td>C-SE</td>
</tr>
<tr>
<td></td>
<td>OptiBond XTR</td>
<td>0-XTR</td>
</tr>
</tbody>
</table>
RESULTS

Microtensile Bond Strength

Two-way ANOVA disclosed statistically significant differences for the main factors adhesive strategy (p = 0.000) and storage time (p = 0.000), as well as their interaction (p = 0.025) (Table 3). The overall mean μTBS of all experimental groups, including the standard deviations, the number of pre-test failures (ptfs), and the number of specimens (n) are detailed in Table 4 and graphically presented in Fig 2. The results of multiple comparisons statistical analysis are also shown in Table 4.

Regarding immediate bond strength, the use of an extra hydrophobic resin coating with or without prior light curing of the universal adhesive did not significantly influence the mean μTBS of the universal adhesives (p > 0.05). Both universal adhesives tested resulted in lower mean μTBSs than the reference two-step self-etch adhesives (p < 0.05). Significant differences in mean μTBS were also not detected between the reference two-step self-etch adhesives (p > 0.05).

Regarding aged bond strength, the application of an extra hydrophobic resin coating increased the aged μTBS of the universal adhesives, but this was only statistically significant when the universal adhesives were light cured first (p < 0.05). The two universal adhesives tested resulted in a lower μTBS after aging than the reference two-step self-etch adhesives (p < 0.05). No significant differences were detected between the reference two-step self-etch adhesives (p > 0.05).

Storage time significantly reduced the mean μTBS of all test groups (p < 0.05), except for light-cured Clearfil Universal Bond with a hydrophobic adhesive layer, which had similar results for both storage times (p > 0.05).

Failure Analysis

Pre-test failures were recorded for all universal adhesive experimental groups, although they were relatively low in number (5.5%). No pre-test failures were recorded for the two reference two-step self-etch adhesives. All pre-test failures occurred during sectioning of the sticks with the diamond saw and these failures occurred adhesively. The results of the light-microscopy failure analysis are graphically presented in Fig 3. Most specimens failed adhesively, and the fewest specimens failed cohesively in dentin, irrespective of adhesive, experimental condition, or storage time. Some representative SEM photomicrographs are illustrated in Fig 4.

### Table 3 Two-way ANOVA for the microtensile bond strength

<table>
<thead>
<tr>
<th>Source</th>
<th>Sum of squares</th>
<th>df</th>
<th>Mean square</th>
<th>F</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>Adhesive strategy</td>
<td>5944.706</td>
<td>7</td>
<td>849.244</td>
<td>173.112</td>
<td>0.000*</td>
</tr>
<tr>
<td>Storage time</td>
<td>1320.489</td>
<td>1</td>
<td>1320.489</td>
<td>107.843</td>
<td>0.000*</td>
</tr>
<tr>
<td>Interaction</td>
<td>211.566</td>
<td>7</td>
<td>30.224</td>
<td>2.468</td>
<td>0.025*</td>
</tr>
</tbody>
</table>

*Statistically significant differences (p < 0.05).

### Table 4 Mean microtensile bond strengths (μTBS in MPa ± SD) of the different experimental groups

<table>
<thead>
<tr>
<th>Experimental groups</th>
<th>Immediate μTBS (24 hrs)</th>
<th>Aged μTBS (6 months)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>MPa ± SD</td>
<td>ptf/n</td>
</tr>
<tr>
<td>CUB</td>
<td>32.42 ± 3.45^AaA</td>
<td>5/45</td>
</tr>
<tr>
<td>CUBnonLC+C-SEbond</td>
<td>33.76 ± 3.64^AbA</td>
<td>3/43</td>
</tr>
<tr>
<td>CUBLC+C-SEbond</td>
<td>34.48 ± 2.78^AbA</td>
<td>2/43</td>
</tr>
<tr>
<td>SBU</td>
<td>34.03 ± 3.69^AbA</td>
<td>3/43</td>
</tr>
<tr>
<td>SBUnonLC+C-SEbond</td>
<td>34.46 ± 3.30^AbA</td>
<td>3/44</td>
</tr>
<tr>
<td>SBULC+C-SEbond</td>
<td>36.38 ± 3.35^AbA</td>
<td>2/44</td>
</tr>
<tr>
<td>C-SE</td>
<td>47.08 ± 1.56^AbA</td>
<td>0/50</td>
</tr>
<tr>
<td>O-XTR</td>
<td>45.38 ± 2.77^AbA</td>
<td>0/50</td>
</tr>
</tbody>
</table>

SD = standard deviation; ptf = pre-test failures (included as 0 MPa); n = total number of specimens (n differs among the experimental groups because teeth differ in size and can reveal different numbers of sticks; 10 teeth per experimental group); same superscript small or capital letter indicates no statistically significant difference in the columns and rows, respectively.
DISCUSSION

In recent years, the application of an extra hydrophobic resin layer has been recommended to improve the adhesion of one-step (self-etch) and universal adhesives to dentin.\(^1,13,20,23,24,29\) In this study, the two universal adhesives Clearfil Universal Bond (Kuraray Noritake) and Single Bond Universal (3M Oral Care) were investigated with the adhesive separately light cured or not prior to the extra application of Clearfil SE Bond bond (Kuraray Noritake). Although higher μTBSs of the universal adhesives to dentin were achieved upon use of the extra hydrophobic layer, only when the universal adhesive was separately light cured before the application of Clearfil SE Bond bond (Kuraray Noritake) did the dentin bond strength significantly improve. When the universal adhesive was not light cured first and thus used as a kind of primer prior to the extra hydrophobic resin layer, no significant increase in bond strength was recorded. Therefore, the hypothesis that the use of an additional hydrophobic adhesive layer applied on a non-light-
Fig 4  SEM photomicrographs of the fracture surfaces (dentin [1] and composite [2] parts) of specimens produced by non-light-cured/light-cured Clearfil Universal Bond and Single Bond Universal followed by application of an extra hydrophobic adhesive layer. Adhesive and mixed failure patterns were revealed. (a) CUB\textsubscript{nonLC}+ C-SE\textsubscript{bond} immediate: A large part of the total surface area of the specimen failed interfacially (adhesive failure), which was characterized by scratch marks remaining from the laboratory-prepared dentin smear layer (arrows). Part of the adhesive resin chipped off during testing. (b) CUB\textsubscript{LC}+ C-SE\textsubscript{bond} aged: Cohesive failure of the adhesive resin, cohesive failure in the composite resin, and interfacial failure (arrows) typically occurred in this specimen (mixed failure). Many small porosities can be observed in the adhesive resin itself (block arrows, composite counterpart). (c) SBU\textsubscript{nonLC}+C-SE\textsubscript{bond} immediate: A small part of the specimen failed near the interface and a large part failed within the adhesive resin (recorded as adhesive failure). (d) SBU\textsubscript{LC}+C-SE\textsubscript{bond} aged: The specimen failed at the interface (adhesive failure). Smooth appearance of the adhesive resin left on the composite side can be observed.
cured universal adhesive would not improve both the immediate and aged bond strength on dentin, was accepted. In previous studies conducted elsewhere, however, significantly higher dentin bond strengths were obtained using universal adhesives (All-Bond Universal, Bisco; Schamburg, IL, USA; Single Bond Universal, 3M Oral Care) applied in self-etch mode when separately light cured prior to the application of an extra hydrophobic resin layer.\(^{20,29}\) In those studies, the application of the universal adhesives was followed by one coat of the hydrophobic resin Heliobond (Ivoclar Vivadent; Schaan, Liechtenstein). The authors attributed the greater bonding effectiveness to the mechanical interfacial properties improved by decreasing the concentration of retained solvents and unreacted monomers in the adhesive layer.\(^{6,20,29}\) and also thanks to the increased thickness of the adhesive layer.\(^{24,29}\)

In the current study, a beneficial effect of the extra hydrophobic resin layer was obtained only when the universal adhesives were separately light cured before the extra layer of Clearfil SE Bond (Kuraray Noritake) was applied and this solely for the aged dentin bond strength. Consequently, the hypothesis that the use of an additional hydrophobic adhesive layer applied on a separately light-cured universal adhesive would not improve the immediate or aged bonding performance to dentin was accepted regarding immediate bond strength, but failed to be accepted regarding aged bond strength. Simplified one-step adhesives that combine the primer function with that of the adhesive resin in multi-step adhesives are generally more hydrophilic and contain more solvent. As a result, these adhesives have been documented to behave as semi-permeable membranes\(^{5,27,34,35}\) and commonly attain lower polymerization conversion rates in part due to remaining solvent.\(^{3,22}\) Thus, these adhesives are more prone to hydrolytic degradation processes, as the resultant adhesive interface is less hydrophobic and thus to a lesser degree impairs water sorption through osmosis from the underlying dentin\(^{5,33,39,40}\) or from the external environment.\(^{24,29,33}\)

Somewhat unexpected in this study was that the extra application of the hydrophobic resin layer on top of the universal adhesive that was solely employed as primer and not separately light cured appeared insufficient to result in a more aging-resistant adhesive interface. Indeed, the highest bond strengths after 6 months of water storage were observed in the experimental group in which the extra hydrophobic resin layer was applied on top of universal adhesives that had been light cured first. Previous laboratory research revealed bonding performance to improve with thickness of the adhesive layer, and increased bond strengths were achieved by applying multiple adhesive coats that were each separately light cured.\(^{1,11}\) Universal adhesives commonly have a rather thin film thickness (< 10 μm), in particular as more solvent evaporation by air blowing/thinning may be indicated because of their higher solvent content.\(^{2}\) Separately light curing the universal adhesives used in self-etch mode before application of the extra layer of Clearfil SE Bond may have thickened the adhesive layer and improved the aging resistance.

Both the universal adhesives applied under the different experimental conditions and the two two-step self-etch reference adhesives presented with significantly lower bond strengths upon 6-month aging than at 24 h (immediate μTBS). The only exception is the universal adhesive Clearfil Universal Bond when it was separately light cured and covered by the extra hydrophobic adhesive layer (CUBLC + C-SEbond): then, statistically non-significantly different bond strengths were recorded at the two time points. Furthermore, in other long-term bonding efficacy studies, Clearfil Universal Bond (Kuraray Noritake)\(^{4,45}\) and Single Bond Universal (3M Oral Care),\(^{4,17,26,29,43}\) when applied in self-etch mode, proved incapable of producing similar μTBS to dentin when the adhesives were applied without an extra hydrophobic adhesive layer, irrespective of specimen aging. Some studies, however, reported otherwise; the application of Clearfil Universal Bond (Kuraray Noritake)\(^{8}\) and Single Bond Universal (3M Oral Care)\(^{19,36,44,46,55}\) in self-etch mode without a hydrophobic adhesive layer resulted in dentin bonding efficacy that was similar initially and after 6-month or 1-year aging. In one study that also investigated the influence of hydrophobic resin coating on bonding effectiveness,\(^{29}\) the authors showed that Single Bond Universal (3M Oral Care) applied in self-etch mode did not result in a significant reduction after 6-month aging when a hydrophobic resin coating was applied; significantly reduced 6-month bond strength was recorded when no extra hydrophobic resin coating was applied. However, in the same study,\(^{29}\) the μTBS after 6 months of aging was not statistically different from that recorded at 24 h for All-Bond Universal (Bisco) with or without an extra hydrophobic coating applied on a separately light-cured universal adhesive used in self-etch mode. The authors concluded that aging stability was material dependent.\(^{29}\) Recently, some clinical studies have indicated a good performance for the universal adhesive All-Bond Universal (Bisco) as well as the two-step self-etch adhesive Optibond XTR (Kerr) in Class II composite restorations after three years of clinical service.\(^{38}\)

Both the universal adhesives Clearfil Universal Bond (Kuraray Noritake) and Single Bond Universal (3M Oral Care) contain the monofunctional monomer HEMA as a hydrophilic monomer (Table 1). HEMA is often added to adhesive formulations (1) to promote wetting of the hydrophilic dentin surface, (2) to facilitate resin infiltration thanks to its hydrophilicity and small size, in particular for etch-and-rinse adhesives/modes, and (3) because of its capacity to act as a solvent for other methacrylate monomers, hereby avoiding phase separation.\(^{21,31,39,41}\) It is a disadvantage that HEMA’s higher hydrophilicity and lower polymerization potential as a monomethacrylate monomer cause HEMA-rich adhesives to absorb more water than HEMA-free/poor adhesives.\(^{16,31}\) Long-term water storage was shown to reduce the tensile strength of adhesives to a degree related to their hydrophilicity.\(^{7,8}\) Interface biodegradation is also induced by activated endogonic matrix metalloproteinases and cathepsins, a process also requiring an aqueous environment.\(^{12,14,28,36}\) In this respect, a water-poor and highly hydrophobic interface that minimizes watersorption is desirable.
The immediate (24 h) and aged (6-month) bonding performance to dentin of the two universal adhesives Clearfil Universal Bond and Single Bond Universal used in this study in self-etch mode did not differ significantly from each other at the two measurement time points. Both adhesives include 10-MDP (10-methacryloyloxydecyl dihydrogen phosphate) as the acidic functional monomer, which, besides creating surface micrometions, also primarily chemically interacts with calcium in hydroxyapatite.\(^{52,53}\) Single Bond Universal (3M Oral Care) also contains a polyalkenoic acid co-polymer, which can also bond chemically to hydroxyapatite.\(^{4,51}\) although it is not clear if the co-polymer is added in a sufficiently high percentage to be chemically interactive. In this respect, the polyalkenoic-acid co-polymer has also been reported to potentially compete with the 10-MDP functional monomer for calcium-bonding sites in hydroxyapatite; it may also inhibit monomer polymerization due to its high molecular weight.\(^{20,44}\) In previous studies, Single Bond Universal (3M Oral Care) was demonstrated to outperform Clearfil Universal Bond (Kuraray Noritake) in terms of bond strength to dentin when both were used in self-etch mode,\(^{4,37}\) which differed from the results of this study. Otherwise, the mean dentin \(\mu\)TBS of Single Bond Universal (3M Oral Care) obtained in this study is in agreement with previous studies, in which the bond strengths ranged from 27.6 to 59.9 MPa depending on the kind of experiment.\(^{4,8,17,20,26,29,32,36,37,43,44,46,55}\)

Favorable findings were recorded for the reference two-step self-etch adhesives Clearfil SE Bond and Optibond XTR; the universal adhesives investigated significantly underperformed the two-step self-etch adhesives. Noteworthy is the significantly higher bonding performance of the reference adhesives for both the immediate and aged \(\mu\)TBS as compared to the universal adhesives applied in all the different adhesive modes. Likewise, no pre-test failures were recorded for the reference adhesives in contrast with all experimental groups of the universal adhesives, although the number of pre-test failures still can be considered relatively low. The bond strength results in other studies that investigated the same universal and reference adhesives tend to agree with the present findings.\(^{8,18,19,26,37}\) The stable bonding effectiveness of the mild two-step self-etch adhesive Clearfil SE Bond (Kuraray Noritake) is believed to be related to the 10-MDP functional monomer, probably along with other properties of primary importance with regard to bonding efficacy and durability, such as efficient polymerization and the application of a separate hydrophobic bonding agent that does not contain solvent but 10-MDP and HEMA.\(^{50,53}\) In contrast to Clearfil SE Bond, the two-step self-etch adhesive Optibond XTR contains GPDM (glycerol phosphate dimethacrylate). The new self-etch adhesive is claimed to feature a primer with enhanced etching ability. Such ability results from the rapid evaporation of acetone which concentrates water and GPDM monomers, thus lowering the \(pH\) from the initial value of 2.4 to 1.6.\(^{45}\) The interfacial chemical interaction of GPDM with hydroxyapatite and dentin has only recently been characterized,\(^{54}\) although the monomer has long been utilized in adhesives. Findings suggested a weak bond between GPDM, hydroxyapatite and unstable calcium salts, although GPDM adsorbed to hydroxyapatite.\(^{46,54}\)

**CONCLUSION**

The bond durability of the two universal adhesives investigated, when applied in self-etch mode, benefited from the application of an extra hydrophilic adhesive layer, but only when the universal adhesive was first separately light cured. Nevertheless, compared to the two reference two-step self-etch adhesives, the universal adhesives investigated revealed lower immediate and aged microtensile bond strengths when applied following the different adhesive strategies. Self-evidently, long-term clinical randomized clinical trials are needed to validate the laboratory bonding results.

**ACKNOWLEDGMENTS**

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**REFERENCES**


Clinical relevance: Universal adhesives applied in self-etch mode on dentin may benefit from an extra hydrophobic adhesive layer when they are light cured first.