Dentin Bond Strengths of Four Adhesion Strategies after Thermal Fatigue and 6-Month Water Storage

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ABSTRACT

Statement of Problem: The stability of current dental adhesives after artificial aging may depend on the adhesion strategy.

Purpose: To evaluate the effect of thermal fatigue and water storage on the dentin microtensile bond strengths (μTBS) of four adhesion strategies.

Materials and Methods: Forty-eight human molars were assigned to four dentin adhesives: FL—OptiBond FL (Kerr Corporation, Orange, CA, USA); SOLO—OptiBond SOLO Plus (Kerr Corporation); XTR—OptiBond XTR (Kerr Corporation); and AIO—OptiBond All-in-One (Kerr Corporation). Teeth were restored with a hybrid composite, and sectioned to obtained bonded beams. For each adhesive, one-third of the central and peripheral beams were assigned to one of three aging conditions: (1) kept in distilled water for 24 hours (*24h*); (2) thermocycled (*TC*) for 20,000 cycles; and (3) stored in distilled water for 6 months (*6M*). Beams were tested in tension mode. Statistical analysis ($p < 0.05$) was computed using Analysis of Variance and Fisher's Least Significant Difference post hoc test.

Results: The highest mean μTBS for *24h*, *TC*, and *6M* were obtained with XTR, but only the *6M* mean μTBS were significantly higher than those of the other three adhesives. For FL, mean μTBS decreased significantly from *24h* to *TC*. For SOLO, mean μTBS remained stable over the three aging conditions. Mean μTBS for AIO decreased significantly from *24h* to *6M*.

Conclusion: The self-etch adhesives XTR and AIO performed similarly or better than the etch-and-rinse adhesives FL and SOLO for all three testing conditions. Their aging stability seems to be material-dependent.

CLINICAL SIGNIFICANCE

The dentin bonding ability of the newest self-etch adhesives has improved compared with other similar materials, including etch-and-rinse adhesives. Adhesives that contain a hydrophobic resin (FL and XTR) tend to be more stable in water storage.

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INTRODUCTION

Current dentin adhesion strategies depend on how the adhesive interacts with the smear layer. Etch-and-rinse adhesives remove the smear layer upon acid-etching, whereas self-etch adhesives make the smear layer permeable without removing it completely.[1]

Whereas multi-bottle etch-and-rinse adhesives involve a separate etching and rinsing step, followed by priming

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and the application of an adhesive or hydrophobic bonding, two-step etch-and-rinse adhesives combine primer and adhesive resin into one solution. Three-step etch-and-rinse adhesives have resulted in better laboratory and clinical performance than two-step etch-and-rinse adhesives.[2,3] The simplification from three- to two-step etch-and-rinse adhesives has some disadvantages, as two-step etch-and-rinse adhesives may need more than one application to achieve a micro-mechanical interlocking of monomers into the collagen-rich etched dentin.[4] Additionally, the lack of a hydrophobic resin coating in two-step etch-and-rinse adhesives may result in degradation of the bonded interface by hydrolysis from fluid transudation through the hybrid layer.[5]

Self-etch adhesives are user-friendlier than etch-and-rinse adhesives because their application time is reduced—no separate acid-etching and no rinsing steps. Self-etch adhesives have become very popular,[6] not only because they are less time-consuming during the restorative procedure, but also because etching and priming are considered technique-sensitive application procedures.[1] Self-etch adhesives consist of non-rinsing acidic monomers dissolved in an aqueous solution, relying on their ability to infiltrate through smear layers to generate a hybrid layer with minerals incorporated.[3] Two-step self-etch adhesives are composed of an acidic primer and a hydrophobic bonding resin, whereas one-step self-etch adhesives do not include the hydrophobic bonding resin, making the latter less effective in vitro and clinically.[7–9] The aggressiveness of self-etch adhesives (i.e., their ability to demineralize dentin and enamel) depends on their pH—mild, moderate, or aggressive self-etch adhesives.[10]

Whereas most bond strength studies only report 24-hour data, other studies have reported bond strengths after thermal fatigue (thermocycling). The in vitro simulation of clinical aging is often performed as an alternative for more time-consuming clinical studies. It has been estimated that 10,000 thermal cycles correspond approximately to 1 year of thermal fatigue in the mouth.[11] The major factor that influences the bonding durability is the hydrolysis of dentin–resin interface components, such as collagen and resin, and subsequent elution of the breakdown products. Hot water may accelerate hydrolysis of non-protected collagen fibers and extract poorly polymerized resin oligomers.[12] Long-term water storage is also an important artificial aging method as it promotes the degradation of the interface by hydrolysis and the plasticization of the polymer matrix,[13] especially if the specimens are aged as individual beams rather than as the entire restored tooth.[12]

Currently, several manufacturers carry different types of dental adhesives, which make the selection very confusing for dental clinicians. Therefore, this study tested the null hypotheses that (1) there are no differences in dentin microtensile bond strengths (μTBS) among four adhesion strategies from the same manufacturer, and that (2) artificial aging does not compromise μTBS of any of the four adhesion strategies.

MATERIALS AND METHODS

Forty-eight intact caries-free sound molars stored in 0.5% chloramine solution for up to 1 month were used in this study. The teeth were examined for structural defects, and left in distilled water for 24 hours (*24h*) at 4°C. All teeth were cleaned with a prophy cup under a slow speed for 15 seconds, and randomly assigned to four groups (listed in Table 1):

1 Group FL—OptiBond FL (Kerr Corporation, Orange, CA, USA), a three-step etch-and-rinse adhesive
2 Group SOLO—OptiBond SOLO Plus (Kerr Corporation), a two-step etch-and-rinse adhesive
3 Group XTR—OptiBond XTR (Kerr Corporation), a two-step self-etch adhesive
4 Group AIO—OptiBond All-in-One (Kerr Corporation), a one-step self-etch adhesive

Middle dentin was exposed by sectioning the crowns parallel to the occlusal surface in a slow-speed diamond saw (IsoMet 1000, Buehler Ltd., Lake Bluff, IL, USA) under water cooling. Dentin was polished with wet 600-grit SiC abrasive paper for 60 seconds to create a

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TABLE 1. Materials batch numbers and composition

OptiBond FL pH primer = 1.8	Gel etchant: 37.5% H_3PO_4, water, fumed silica LOT: 3353342	FL primer: HEMA, GPDM, MMEP, water, ethanol, photoinitiator (CQ), BHT LOT: 3549622	FL adhesive: Bis-GMA, HEMA, GPDM, GDMA, photoinitiator (CQ), ODMAB, fillers (fumed SiO_2, barium aluminoborosilicate, Na2SiF6), coupling factor A174 LOT: 3538016
OptiBond SOLO Plus pH = 2.1	Gel etchant: 37.5% H_3PO_4, water, fumed silica LOT: 3353342	Bis-GMA, HEMA, GPDM, water, ethanol, barium aluminoborosilicate glass, fumed silica (silicon dioxide), sodium hexafluorosilicate, photoinitiator (CQ) LOT: 3661965	
OptiBond XTR pH primer = 2.4 prior to application; drops to 1.6 upon application to tooth structure	XTR primer: Acetone, water, ethanol, HEMA, photoinitiator (CQ), GPDM LOT 3594446	XTR adhesive: Ethanol, HEMA, sodium hexafluorosilicate, MEHQ; nano-silica, barium; photoinitiator (CQ) LOT 3594446	
OptiBond All-in-One pH = 2.5–3.0	GPDM, HEMA, GDMA, Bis-GMA, water, ethanol, acetone, photoinitiator (CQ), silica, sodium hexafluorosilicate LOT: LD01072		
Filtek Z250	Bis-EMA, TEGDMA, UDMA, zirconium, silica LOT: N197646		

bis-GMA = bisphenol A diglycidyl methacrylate; Bis-EMA(6)1 (Bisphenol A polyethylene glycol diether dimethacrylate); BHT = butylhydroxytoluene; CQ = camphorquinone; DMA = dimethacrylates; GDMA = glycerol dimethacrylate; GPDM = glycerol phosphate dimethacrylate; HEMA = 2-hydroxyethyl methacrylate; MAC-10 = 11-methacryloyloxy-1,1'-undecanedicarboxylic acid; 10-MDP = 10-methacryloyloxydecyl dihydrogen phosphate; MEHQ = 4-methoxyphenolMono(2-methacryloyloxy)ethyl phthalate; ODMAB = 2-(ethylhexyl)-4-(dimethylamino)benzoate; TEGDMA = triethyleneglycol-dimethacrylate; 4-META = 4-methacryloyloxyethyl trimellitate anhydride; H_3PO_4 = phosphoric acid.

Dr. Eugene Qian, Principal Scientist, Kerr Corporation, personal communication, July 6, 2011.

standardized smear layer.[13] After the application and light-curing of the adhesives (Table 2) with the Elipar S10 Curing Light (3M ESPE, St. Paul, MN, USA), crowns were restored with Filtek Z250 hybrid composite resin (3M ESPE, shade A2) in three increments of 2 mm each. Each increment was irradiated for 40 seconds. After the build-up was completed, a 3×3 mm^2 square was painted in the central area of the composite occlusal surface with a colored permanent marker. The peripheral area was painted with a different color to allow for the selection of central and peripheral bonded beams. The crowns were then sectioned automatically under water irrigation with a slow-speed diamond saw (Accutom 50, Struers A/S, Ballerup, Denmark) in X and Y directions to obtain beams with a cross-section of 0.5 ± 0.2 mm^2. For each adhesive, the beams were assigned into three aging conditions—one-third of the central and peripheral beams from each restored tooth were kept in distilled water for 24 hours at 37°C (*24h*); one-third of the central and peripheral beams were thermocycled for 20,000 cycles (AraLab REFRI 200E, AraLab, Rio de Mouro, Portugal) between water baths held at 5 and 55°C, with a dwell time of 30 seconds (*TC*); and the remaining one-third was stored in distilled water containing 0.4% sodium azide (pH = 7.0),[14] for 6 months at 37°C.[15] The storage solution was changed every week.[16] The beams were individually attached to a stainless steel notched Geraldeli's jig[17] using cyanoacrylate glue (Zapit, Dental Ventures of America, Corona, CA, USA), and then submitted to a tension load using a Shimadzu Autograph (Shimadzu AG-IS, Tokyo, Japan) universal testing machine at 1 mm/minute crosshead speed. A Storm Digital Caliper (Pontoglio, Brescia, Italy) with an accuracy of 0.01 mm was used to measure the sides of the bonding interface and calculate the bonding area in mm^2. The load at fracture and the bonding surface area of the specimen

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TABLE 2. Instructions for use

Adhesives	Classification	Manufacturer's instructions
OptiBond FL	Three-step etch-and-rinse adhesive, light-cured	Etch dentin/enamel with Kerr Gel Etchant for 15 seconds; rinse thoroughly for 15 seconds; air-dry for 3 seconds (do not desiccate); apply primer with brushing motion for 15 seconds; air-dry for 5 seconds; using same applicator, apply adhesive with light brushing motion for 15 seconds; air thin for 3 seconds; light-cure for 20 seconds
OptiBond SOLO Plus	Two-step etch-and-rinse adhesive, light-cured	Etch dentin/enamel with Kerr Gel Etchant for 15 seconds; rinse thoroughly until all acid is removed; dry lightly (do not desiccate); apply OptiBond Solo Plus with light brushing motion for 15 seconds; air thin for 3 seconds; light-cure for 20 seconds
OptiBond XTR	Two-step self-etch adhesive, light-cured	Prepare cavity; pumice clean unprepared tooth structure with a fluoride-free cleaning paste; rinse thoroughly with water spray and air-dry; apply primer to the enamel/dentin surface using the disposable applicator brush; scrub the surface with a brushing motion for 20 seconds; air thin for 5 seconds with medium air pressure; shake adhesive bottle briefly; apply adhesive to the enamel/dentin surface with light brushing motion for 15 seconds; air thin for 5 seconds; light-cure for 10 seconds
OptiBond All-in-One	One-step self-etch adhesive, light-cured	For bottle: dispense 2–3 drops of OptiBond All-In-One adhesive into a clean well. For Unidose container: open the container and insert the applicator into the container to saturate the applicator tip.
		Apply a generous amount of OptiBond All-In-One adhesive; scrub the surface with a brushing motion for 20 seconds; apply a second application of OptiBond All-In-One adhesive with brushing motion for 20 seconds; thoroughly dry the adhesive gentle air first and then medium air for at least 5 seconds; light-cure for 10 seconds

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were registered, and microtensile bond strengths were calculated in MPa. The fractures were analyzed by two observers under a stereo microscope (Leica MZ6, Leica Microsystems AG, Heerbrugg, Switzerland) at ×20. The mode of failure was classified as adhesive, mixed, and cohesive. Failures were considered adhesive when they occurred at the dentin–adhesive interface; they were of cohesive nature when the failure occurred in dentin; and of mixed nature when there was composite and dentin at the interface.

Statistical analysis was computed with PASW Statistics 18 (SPSS Inc., Chicago, IL, USA) using Analysis of Variance and Fisher's Least Significant Difference post hoc multiple comparison tests to analyze mean μTBS. For each of the adhesives, the beams from each tooth were averaged to represent the experimental unit (12 teeth per adhesive for each of the three aging conditions).

RESULTS

The mean microtensile bond strengths, respective standard deviations, and statistical differences are displayed in Table 3.

By Aging Method

At *24h*, the highest mean μTBS were obtained with XTR, but not statistically different from those of AIO

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TABLE 3. Mean μTBS ± SD (MPa) and statistical differences

	24h	TC	6 m
OptiBond FL	$57.2 \pm 11.3^{A,*,\#}$	$50.7 \pm 13.2^{a,\#}$	$62.2 \pm 16.0^{\alpha,*}$
OptiBond SOLO Plus	$62.6 \pm 14.4^{A,B,*}$	$55.7 \pm 9.8^{a,b,*}$	$57.5 \pm 13.2^{\alpha,*}$
OptiBond XTR	$70.0 \pm 14.2^{B,*}$	$65.1 \pm 10.2^{c,*}$	$81.6 \pm 18.2^{\beta,\#}$
OptiBond All-in-One	$63.6 \pm 16.1^{A,B,\#}$	$59.7 \pm 11.7^{a,b,c,*,\#}$	$50.1 \pm 17.1^{\alpha,*}$

Columns: Means with the same superscript letter are not statistically different at $p < 0.05$.

Rows: Means with the same superscript symbol are not statistically different at $p < 0.05$.

($p > 0.272$) and SOLO ($p > 0.204$). FL resulted in the lowest mean μTBS, which were statistically lower than those of XTR at $p < 0.031$, but statistically similar to those of AIO ($p > 0.270$) and SOLO ($p > 0.353$). AIO and SOLO resulted in virtually similar mean μTBS ($p > 0.858$).

For *TC*, the highest mean μTBS were obtained with XTR, but not statistically different from those of AIO ($p > 0.248$). SOLO resulted in statistically lower mean μTBS than those of XTR ($p < 0.049$). FL resulted in the lowest mean μTBS, which were statistically lower than those of XTR ($p < 0.003$), but statistically similar to those of AIO ($p > 0.060$) and SOLO ($p > 0.289$). AIO and SOLO resulted in statistically similar mean μTBS ($p > 0.289$).

At *6M*, the highest mean μTBS were obtained with XTR, which were statistically different from those of AIO ($p < 0.0001$), SOLO ($p < 0.001$), and FL ($p < 0.005$). FL resulted in the lowest mean μTBS, which were statistically similar to those of AIO ($p > 0.073$) and SOLO ($p > 0.477$). AIO and SOLO resulted in statistically similar mean μTBS ($p > 0.269$).

By Adhesive

For each adhesive, there was no statistical difference between mean μTBS at *24h* versus mean μTBS after *TC* (FL, $p > 0.257$; SOLO, $p > 0.193$; XTR, $p > 0.414$; AIO, $p > 0.530$).

FL resulted in increased mean μTBS at *6M* compared with *TC* ($p < 0.047$), but not statistically different from the mean μTBS at *24h* ($p > 0.369$).

For SOLO, the mean μTBS at *24h* and after *TC* were not statistically different from mean μTBS at *6M* (*24h*, $p > 0.332$; *TC*, $p > 0.734$).

XTR resulted in higher mean μTBS at *6M* than at *TC* ($p < 0.009$). The *p*-value between mean μTBS at *6M* and at *24h* was close to significance ($p = 0.058$).

AIO resulted in a significant decrease in mean μTBS from *24h* to *6M* ($p < 0.036$), but not statistically different from mean μTBS after *TC*.

There were no pretesting failures. Over 69% of the failures were of adhesive nature, equally distributed among the four adhesives.

DISCUSSION

We failed to accept the first null hypothesis as there were statistical differences in dentin μTBS among the four adhesion strategies from the same manufacturer. The second null hypothesis has to be rejected as some adhesives were affected by the 6-month water storage.

All four adhesives used in this study contain glycerol phosphate dimethacrylate or GPDM, a phosphate monomer that has been used in dentin bonding for over 50 years. In 1952, it was reported that a resin containing GPDM stained the "altered" dentin immediately below the filling material. This was the first historical report of changes in dentin promoted by an acidic monomer, and may be considered the precursor of the hybrid layer concept.[18,19] In 1956, a resin containing GPDM was used to bond to hydrochloric acid-etched dentin.[20]

For etch-and-rinse adhesives, bond strength is derived both from resin tag and hybrid layer formation,[21] with resin tags being responsible for an estimated one-third of the bond strengths.[22] However, for the recent self-etch adhesives, chemical bonding has been shown

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to play a more important role.[23,24] Mild self-etch adhesives rely on a combination of chemical adhesion to hydroxyapatite with some micro-mechanical interlocking that does not depend much on resin tag formation. This twofold adhesive mechanism may be advantageous for bonding durability.[12] Taking into consideration that different bonding strategies (i.e., etch-and-rinse and self-etch) are influenced by chemical bonding and by micromechanical interlocking in different degrees, variation in regional characteristics of the substrate may also affect the two bonding strategies differently. With this in mind, we divided the bonded beams from each tooth in peripheral beams and central beams to avoid any bias in the μTBS distribution caused by regional differences. In fact, there is some controversy as to whether beams obtained from the peripheral area of the restored tooth result in decreased or increased μTBS. Whereas one study reported lower μTBS for peripheral specimens than for centrally located specimens,[25] another study reported that μTBS to "periphery" dentin is higher than for the "center" specimens.[26]

An in vitro study reported that the dentin bond strengths of FL are quite stable after 20,000 thermal cycles, which is in agreement with our study.[27] More recently, the same research group reported 1-week mean μTBS of 53.2 MPa for central beams versus 65.1 MPa for beams from the periphery,[26] which are very similar (on average) to the mean μTBS measured in the present study for FL. Likewise, Heintze and colleagues measured a mean μTBS of 56.7 MPa with FL at 8 hours.[28] The *24h* versus *TC* mean μTBS for FL in our study were not only within the range of values reported in the literature, but also statistically similar to those of AIO and SOLO for either testing period. The *6M* mean μTBS for FL were higher than those obtained with *24h* and *TC*. Fontes and colleagues[29] also reported the highest mean μTBS with FL after 1 year of water storage. The experimental methodology was similar to that in our study, as authors used 0.5-mm^2 cross-section beams stored in distilled water.[29] In resin systems, the monomer conversion continues after the polymerization process, as free radicals continue to propagate and to terminate independently of the water storage condition,[30] which may explain our findings.

We hypothesize that the hardening of FL due to continued free radical polymerization might have superseded the plasticization process. We did not anticipate, however, that the mean μTBS for FL would be statistically lower than those measured for XTR for all the testing periods, as FL has been considered by many experts as one of the references for all adhesives.[1,12,31,32]

For OptiBond SOLO Plus, studies have reported a variety of bond strengths, which may be a consequence of the different testing conditions. Sadek and colleagues[33] obtained 38–40 MPa with microtensile bond testing, but used beams with a bonded area of 0.9×0.9 mm^2, whereas the bonded area of our specimens measured an average 0.5×0.5 mm^2. There is an inverse relationship between microtensile bond strength and bonded surface area.[34] Teixeira and Chain[35] obtained a 24-hour mean shear bond strength of 30.7 MPa, which was not statistically different from the mean that they obtained with Clearfil SE Bond (CSE, Kuraray America Inc., Houston, TX, USA), one of the references in several adhesion studies.[36–40] OptiBond SOLO, the predecessor of OptiBond SOLO Plus, provided good clinical retention rate (69%) in non-carious class V lesions at 8 years as opposed to 59% measured for Prime & Bond 2.1.[41] Additionally, OptiBond SOLO bonds equally to superficial and deep dentin.[42] There was a tendency in our study to decreased mean μTBS for SOLO after aging (*TC* and *6M*); however, not statistically different from the *24h* measurements. This tendency has been reported before for OptiBond SOLO after thermocycling[43] and after water storage.[44,45]

XTR, a two-step mild self-etch adhesive, is very recent; therefore, there are not many dentin bond strength studies available. Walter and colleagues[46] reported mean shear bond strengths at *24h* of 45.1 MPa for XTR and 33.3 MPa for FL, the three-step etch-and-rinse adhesive. Similarly, Campillo-Funollet and colleagues[47] obtained a mean shear bond strength of 56.8 MPa at *24h* for XTR and 49.4 MPa for FL. The results of both studies are in agreement with the findings of our study, as XTR has resulted in higher dentin bond strengths

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than those of FL, a proven etch-and-rinse adhesive. XTR was not negatively affected by the aging conditions in the present study. Bui and colleagues[48] reported no statistical differences among the immediate mean shear bond strength of XTR comparing to mean shear bond strength after thermocycling (5,000 cycles) and after 1-month water storage. Moreover, the relatively high bond strengths obtained with XTR are in the same magnitude of those obtained in other studies with the two-step self-etch adhesive CSE, which is now considered the golden standard for all self-etch adhesives both in vitro and clinically.[36–40,46,49,50]

The in vitro and clinical success of mild self-etch adhesives might be a result of two factors: (1) their chemical composition, which generally includes monomers that form chemical bonds with hydroxyapatite (HAp)[51]; (2) and the presence of a hydrophobic bonding layer.[8,9] The tendency for high bond strengths with XTR suggests that this adhesive may posses an intrinsic ability to bond chemically to dentin, in the same line as CSE, a 10-MDP-containing adhesive. The pH of XTR primer is similar to that of CSE primer. Whereas CSE primer has a pH between 1.76 and 2.0,[52,53] the pH of XTR primer is 2.4 prior to application, but drops instantaneously to 1.6 upon application to tooth structure (Dr. Eugene Qian, Principal Scientist, Kerr Corporation, personal communication, July 6, 2011). The bonding ability of CSE is a result, in part, of the Ca-10-MDP salt being one of the most hydrolytically stable salts.[54] According to the adhesion–decalcification concept,[23] the less soluble the calcium salt of the acidic molecule is, the more intense and stable the molecular adhesion to an HAp-based substrate. MDP is adsorbed onto HAp in a regularly layered structure at the HAp surface (nano-interaction),[23] and at the same time decalcifies HAp.[55] The interaction of other current acidic monomers, such as 4-MET and Phenyl-P, with HAp has been well documented.[23] However, it is not yet known whether GPDM, the phosphate monomer in XTR, has the same potential for stable chemical bonding with HAp. We speculate that a similar chemical interaction between GPDM and HAp might occur, and be responsible for the high bond strengths obtained with both AIO and XTR in the present study. Only clinical studies with XTR will shed some light in the stability of the bonds between GPDM and HAp, as AIO, being a one-step self-etch adhesive, may be more prone to interfacial degradation with time than XTR.

An SEM morphologic analysis of XTR–dentin interface by Qian and colleagues[56] revealed a narrow but well-defined hybrid layer, with long resin tags. However, the role of resin tags on the bonding mechanism of self-etch adhesives is debatable. The morphology, length, and adaptation of resin tags are only indicative of the wetting ability of the respective hydrophilic monomers,[57] and tags have to be firmly bonded to tubules wall to provide retention. Mild self-etch partially dissolve the smear layer, especially if a thick smear layer is present, leading to a narrow hybrid layer and a low resin tag density.[58] A recent in vitro study suggested no influence of resin tags on bond strength,[59] as microtensile bond strengths decreased with or without the presence of resin tags after thermocycling.

There are not abundant bond strength studies with OptiBond All-in-One. AIO bonds equally well to dentin independently of tubule orientation and depth.[60] Kimmes and colleagues[61] measured 26.2 MPa in shear mode with AIO when applied for the time recommended by the manufacturer, which were statistically similar to the shear bond strengths obtained with CSE (29.1 MPa). The similarity in bond strengths between these two adhesives may be a sign that AIO has good chemical bonding potential to dentin. A more recent study[7] found statistically higher dentin μTBS for FL (38.1 MPa) than those for AIO (23.2 MPa), which is in disagreement with the μTBS obtained in our study. This difference may be explained by the operator variability. This particular research group[7] has consistently reported excellent in vitro and clinical behavior with FL, which may be a consequence of their optimal handling of the material.[26,38] In fact, FL has been shown to be more prone to operator variability than other adhesives, such as OptiBond SOLO.[62] Six-month water storage resulted in decreased mean μTBS for AIO compared with those of *24h*. Itoh and colleagues[63] concluded that mean μTBS for AIO were significantly affected by the storage period due to water

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sorption. The authors reported mean μTBS of 47.6 MPa at *24h* that decreased to 39.1 MPa after 1 year of water storage.[63] However, the 6-month mean μTBS for AIO were not significantly different from the *24h* mean μTBS,[63] which is not in agreement with the findings in our study.

The mean μTBS obtained in our study with AIO and SOLO were slightly higher than those reported in other studies, which may be explained by the size of the bonded area.[34] This may be due to the distribution of defects in the material because a larger specimen probably contains many more defects compared with smaller specimens.[34]

The *6M* mean μTBS were higher for the adhesives that have a separate hydrophobic bonding layer (FL and XTR). This is in agreement with a previous study in which FL and CSE maintained higher bond strengths after 6-month and 12-month water storage comparing to the adhesives that combined primer and bonding resin in one solution.[26] Solvents and hydrophilic monomers allow water permeation into the polymerized adhesive.[64] The hydrophilic monomers absorb more water due to their polarity, affecting the mechanical stability of the resins and accelerating the degradation process.[64,65] This water sorption, solubility, and diffusion coefficients are material-dependent.[65] All the adhesives in our study have the same monomer (GPDM) and polar solvents; therefore, the application of a layer of non-solvated hydrophobic monomers may have limited the diffusion of water throughout the hybrid layer[66] and may reduce the faster release of unreacted monomers from the resin–dentin interface.[16] This hydrophobic resin layer may be responsible for low sensitivity to water degradation.[8] This is more relevant when dentin is directly exposed to water without the surrounding resin-enamel protecting barrier.[45]

Whereas for some adhesives thermal fatigue for 20,000 cycles does not affect their dentin bond strengths,[27] the susceptibility of other adhesives to thermal fatigue depends on the specific composition of each adhesive.[67] Although thermocycling may be considered an initial screening method that induces hydrolysis of non-protected collagen fibers and extract poorly polymerized resin oligomers from the interface,[12] long-term water storage may correlate better with clinical behavior. In fact, Heintze and colleagues[28] reported that μTBS of adhesive systems after water storage for 6 months showed a good correlation with marginal discoloration in short-term clinical Class V restorations. Some concerns exist with regard to the change of solution in long-term storage tests. A faster resin–dentin interface degradation is induced when the solution is changed frequently.[16] The time needed to saturate the storage solution depends on the number of specimens per volume. However, durability studies often do not report the amount of water used for storage nor the number of specimens placed within the vials. In our study, an average of seven beams were stored in glass vials containing 20 mL of distilled water. Skovron and colleagues[16] stored each beam individually in 10 mL vials.

Based on the existing literature, simplification of adhesives systems results in loss of bonding effectiveness, although some two-step self-etch approach the golden standard.[12] With this in mind, and to understand how XTR interacts with dentin, future studies that include the characterization of the chemical interaction of GPDM with HAp, and clinical studies of the same materials used in this project, are currently being planned.

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