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Microtensile bond strength and sealing efficiency of all-in-one self-etching adhesives

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This study aimed to evaluate the microtensile bond strength and amount of microleakage with self-etch adhesives. For the bond strength study, 50 human third molars were randomly assigned to five groups according to the adhesive system used. In each group, half of the specimens were subjected to the microtensile test immediately after the bonding procedures and tooth sectioning, while the remaining half were subjected to the test after storage in distilled water for 3 months. For the microleakage study, following the preparation and restoration of class V cavities, 70 human molars were divided into two groups for the five adhesives; the specimens in one group were stored in distilled water for 24 h and those in the other group were stored for 3 months. In both groups, the teeth were sectioned and evaluated for dye penetration after the storage time. After 24 h, the microtensile bond strengths in descending order were as follows: Clearfil SE Bond > G Aenial Bond > Optibond All-in-One (AiO) > Adper Prompt L Pop (hereafter L Pop) > Futurabond M. After 3 months, the μ TBS of all five bonding agents was decreased, although the decrease was significant only for L Pop, which showed the highest scores for leakage around the enamel margins, whereas Futurabond M showed the highest scores for leakage around the gingival margins. After 3 months, the microleakages scores significantly increased for G Aenial Bond, Futurabond M and OptibondAiO.

Keywords: bond strength durability; all in one self-etching adhesives; sealing effect

Introduction

Simplified adhesives, often referred to as all-in-one self-etch adhesives, have gained considerable market share [1] and popularity among dentists since their recent introduction, considering they are available as a single solution containing etchant, primer, and bonding agent held together by both hydrophilic and hydrophobic monomers with a relatively high concentration of solvent.[1,2]

In contemporary adhesives, hydrophilic resin monomers are dissolved in volatile solvents such as acetone and ethanol, which aid in displacing water from the dentinal surface and facilitate resin monomer penetration into the collagen network,[3] increasing bond strength. Water is also essential as an ionization medium to facilitate self-etching activity in self-etch adhesives. Elimination of all solvents and water from the adhesive is essential before light curing to prevent adverse effects on monomer polymerization.[4] This is achieved by allowing an evaporation time between resin application and polymerization. Residual solvent in the adhesive resin or water in the hybrid layer because of high hydrophilic content of the adhesive or inappropriate water evaporation leads to pore formation in the cured adhesive,[5] which can compromise restoration longevity.[6]

Bond durability and the hermetic coating is essential for long-term restoration success, because a poor bond weakens adhesion and leads to gap formation at the tooth-restoration interface.[7] Microleakage predisposes the tooth to post-operative sensitivity, secondary caries and eventual treatment failure.[8]

Bond strength decrease in hydrophilic resin primarily occurs during the first month of water storage *in vitro*. [9] Reportedly, the bond strength curve decreases after 3–6 months of water storage, although it does not decrease to zero; moreover, bond strength is retained after long-term water storage.[10] This decrease may result from water movement within the hybrid layer [11] or hydrolysis of collagen fibrils not encapsulated by resin monomers at the base of the hybrid layer.[12,13]

Taken together, adhesive systems require solvents and hydrophilic monomers to produce a strong bond at the moist dentin surface. However, the solvent and hydrophilic content of the adhesive causes gradual deterioration of adhesives at tooth interfaces. Therefore, this study aimed to investigate the effectiveness of four all-in-one self-etch adhesives with varied hydrophilicity and solvents and one two-step self-etch adhesive (as a control) in terms of microtensile bond strength and microleakage

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after distilled water storage for 24 h and 3 months. The null hypotheses were as follows.

- (1) The solvent content of adhesives influences initial μ TBS values and leakage scores.
- (2) Despite a more user-friendly technique simplified adhesives tend to display an inferior bond strength compared with multistep adhesives.
- (3) Interface deterioration initiates after 3 months of water storage, with decreasing μ TBS values and/or increasing leakage scores.

Materials and methods

Microtensile bond strength

Fifty caries- and defect-free human third molars were obtained from patients requiring extraction. The teeth were disinfected in 0.5% chloramin T (Merck KGaA 64271 Darmstadt, Germany), stored in distilled water at 4 °C and used within 6 months after extraction. The occlusal enamel was horizontally sectioned 1 mm below the dentinoenamel junction (DEJ) using a slow-speed diamond saw (Isomet 1000, Buehler, Lake Bluff, IL, USA). A light microscope (Olympus SZ61, Munster, Germany) was used at 20 \times magnification to ensure accurate occlusal enamel removal. The exposed dentin surfaces were further polished on wet 600-grit SiC paper for 60 s to create a standardized smear layer.

The teeth were then randomized into five groups ($n = 10$), which were randomly assigned to one of the five adhesives being tested. All adhesive systems were applied strictly according to the manufacturer's instructions (Table 1). After application, a composite core build-up was performed using a microhybrid resin composite in three layers of maximum 2-mm thickness to a height of 5 mm (Filtek Z 250, Shade A2, (3M ESPE, St.Paul, USA). Each increment was light-cured with a quartz-tungsten halogen curing unit (Bisco VIP dental curing light, Schumburg, USA) for 40 s. The light intensity output was monitored using a dental radiometer (Hilux Ledmax Light Curing Meter, Benlioğlu Dental Inc, Ankara, Turkey) and was at least 600 Mw/cm². The restored teeth were stored in water at 37 °C for 24 h and longitudinally cut into five or six 1-mm-thick and 10-mm-long sections perpendicular to the tooth-adhesive interface, using a slow-speed diamond saw under water irrigation. The sections were left partially attached to the tooth, which was then rotated 90 degrees and sectioned again to obtain 1-mm²-thick (± 0.2 mm²) and 10-mm-long sticks. Specimens that failed during sectioning were excluded from statistical analyses. All bonding procedures were performed at room temperature by a single operator. The intact sticks were then randomized into two subgroups; specimens in one group were stored in distilled water at

37 °C for 24 h and subjected to bond strength testing, while those in the other group were stored in distilled water at 37 °C for 3 months before being loaded to failure. After the storage period, the specimens were attached to a modified device for microtensile testing with cyanoacrylate resin (Zapit Dental ventures of North America, Corona, CA, USA) and subjected to a tensile force until failure in a universal testing machine (Micro Tensile Tester BISCO Inc, Schaumburg, IL, USA) at a crosshead speed of 0.5 mm/min. The failure load was recorded for each specimen. The failure mode was evaluated at 30 \times (Olympus SZ61) and classified as cohesive within dentin, cohesive within composite resin and adhesive/mixed (failure at resin-dentin interface or concomitant cohesive failure of the neighbouring substrates).[11]

The mean μ TBS values after 24 h and 3 months were calculated for each adhesive. The distribution of μ TBS data was checked for normality using the Kolmogorov-Smirnov test and statistically analysed using one-way ANOVA to examine the effects of adhesive type and testing time on bond strength. When significant differences were found among adhesives, they were compared using Tukey's HSD post-hoc test, with $p < 0.05$ considered statistically significant.

Microleakage

Seventy caries- and defect-free human molars were collected after obtaining patient consent. The teeth were disinfected in 0.5% chloramin T for a week, stored in distilled water at 4 °C, and used within 6 months after extraction. Standard class V buccal cavities were centred on the cement-enamel junction and were approximately 2-mm deep, 3-mm high and 5-mm wide. Maintaining 90° cavosurface angles at all cavity margins produced a slightly divergent preparation with no deliberate mechanical retention.

Each bonding resin was applied on the dentin surface according to the manufacturer's instructions ($n = 7$; Table 1). Following adhesive treatment (Filtek Z 250), a composite core was built with light activation of individual increments for 40 s (Bisco VIP).

Soft-Lex disks (3M ESPE) were used for finishing and polishing. The bonded specimens were stored in distilled water at 37 °C for 24 h before being subjected to 500 thermocycles between water baths at 5 °C and 55 °C. The specimens in each group were tested after 24 h or after water storage for 3 months.

After the storage time, the tooth apices were sealed with amalgam (YDA Amalgam Alloy Capsules, Spain), and all tooth surfaces, except a 1-mm-wide zone around the restoration margin, were sealed with nail varnish. Subsequently, they were immersed in a 0.5% methylene blue dye solution for 24 h. The teeth were then rinsed under tap water and dried. Each tooth was buccolingually

Table 1. Ingredients, manufacturers, application technique and lot number of dentin bonding systems used in this study.

| Dentin bonding agents | Manufacturer | Ingredients | Application | Lot number |
|-----------------------|--|--|--|------------|
| Optibond All-In-One | KERR Kerr Corporation Orange, CA | Monomers –Glycerol phosphate dimethacrylate (GPDDM) Difunctional methacrylate monomers, HEMA Solvents – water, acetone and ethanol Photo-initiator – camphorquinone Fillers – three nano-sized fillers Fluoride-releasing fillers – sodium hexafluoroarsenate and ytterbium fluoride | Apply to tooth for 20 s with agitation twice Repeat this procedure twice Light-cure for 10 s | 2956948 |
| Clearfil SE Bond | KURARAY Medical Okuyama, Japan | Primer: MDP, HEMA, hydrophilic dimethacrylate, N-Diethenol p-toluidine, water Bond: MDP, BisGMA, HEMA, hydrophobic dimethacrylate, CQ, N, N-Diethenol p-toluidine, Silanated colloidal silica | Apply primer 20 s. Mild air stream. Apply bond. Gentle air stream. Light cure 10 s. | 41205 |
| Futurabond M | VOCO Cuxhaven Germany | Urethandimethacrylate, HEMA Acetone Acidic adhesive monomer 2-hydroxyethylmethacrylate Catalyst | Apply adhesive (scrubbing) 20 s. Air blow. Light cure 20 s. | 1015233 |
| G-Aenial Bond | GC CORPORATION Tokyo, Japan | Phosphorylated methacrylates 4-Methacryloxyethyltrimellitate anhydride (4-META) Triethylene glycol dimethacrylates (TEGMA) Urethane dimethacrylates (UDMA) Acetone, water | Apply one coat of adhesive, leave undisturbed for 10 s Air dry vigorously Light-cure for 10 s | 1011181 |
| Adper Prompt L-Pop | 3M ESPE St Paul, MN, USA | HEMA phosphates, HEMA, bis-GMA, modified polyalkenoic acid, water, photoinitiator | Scrub first coat 15 s. Gently air dry. Second coat application (scrubbing) 15 s. Gently air dry. Light-cure 10 s. | 387690 |

BisGMA = 2,2-bis(4-(2-hydroxyloxypropoxy)phenyl)propane; HEMA = 2-hydroxyethylmethacrylate.

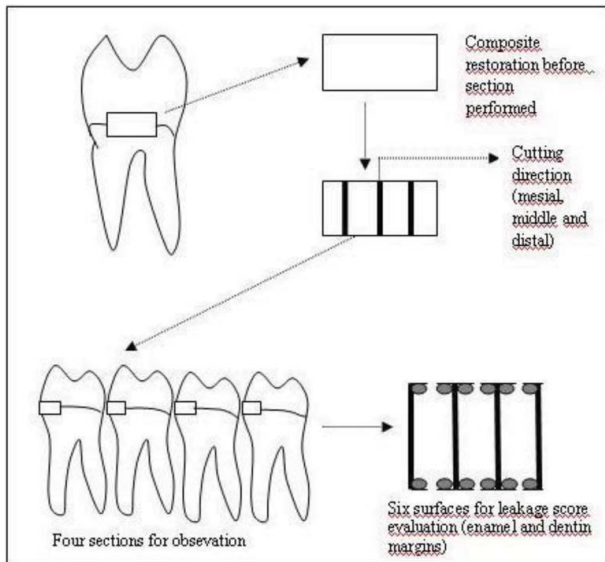


Figure 1. Schematic illustration of cutting restorations on the teeth and methodology of microleakage evaluation. Adapted from Demirci et al. [14] with permission from John Wiley and Sons by License Number: 3559240332458.

sectioned into 0.8-mm-thick sections with a slow-speed diamond saw under water cooling (Isomet 1000) to evaluate dye penetration.

On each restoration, three cuts (mesial, middle, and distal) were prepared longitudinally in the buccolingual direction using a diamond saw mounted on a cutting machine (Isomet 1000; Figure 1). These preparations yielded six evaluating surfaces (four sections) for each restoration, with a total of 420 viewing surfaces. Each specimen allowed one measure in enamel and one in dentin, with a total of 840 measures. The sections were observed under a stereomicroscope (Olympus SZ61, Olympus Corporation, Tokyo, Japan) at 40 \times magnification, and microleakage at the enamel and gingival margins in each section was evaluated by two independent operators using the following scoring system:

- 0: no dye penetration,
- 1: dye penetration up to DEJ in the enamel margin or up to one-third of the full gingival margin wall length,
- 2: dye penetration beyond DEJ and up to two-thirds of the full enamel margin wall length or between

one- and two-thirds of the full cervical margin length,

- 3: dye penetration beyond two-thirds of the full enamel or gingival wall length, without axial wall involvement,
- 4: extensive dye penetration with axial wall involvement.

Borderline cases were decided by consensus among observers. Statistical analyses were conducted using the Kruskal–Wallis test to determine statistically significant differences in leakage at the enamel and gingival margins among groups for each adhesive and among the five adhesives within each group. If a significant difference was observed at any margin, Dunn’s multiple comparison test was performed. Intergroup comparisons (enamel vs. gingival margins) were conducted using the Wilcoxon signed-rank test. All statistical tests were performed at $p < 0.05$.

Results and discussion

Microtensile bond strength

The mean μ TBS values obtained with the five bonding agents after 24 h and 3 months are shown in Table 2. Pre-testing failures were excluded from analysis. One-way ANOVA revealed statistically significant differences for adhesive type ($p < 0.05$), with significant differences in μ TBS values among all bonding agents at 24 h: Clearfil SE Bond (43.9 ± 6.8 MPa) > G Aenial Bond (35.3 ± 6.2 MPa) > Optibond All-In-One (AiO; 30.4 ± 5.3 MPa) > Adper Prompt L Pop (L Pop; 26.2 ± 6.2 MPa) > Futurabond M (18.4 ± 3.8 MPa). After 3 months, the μ TBS values were significantly different among all except between L Pop and Futurabond M: Clearfil SE Bond (42.1 ± 6.6 MPa) > G Aenial Bond (32.7 ± 5.9 MPa) > Optibond All-In-One (28.1 ± 5.6 MPa) > L Pop (21.2 ± 5.8 MPa) > Futurabond M (17.0 ± 3.5 MPa).

The μ TBS values of all adhesives decreased slightly over time, with only L Pop showing a significant decrease after 3 months.

The failure modes observed under light microscopy are presented in Table 3. The majority of failure modes for each adhesive were adhesive/mixed after 24 h and 3 months. Although the overall failure percentage was markedly lower than the adhesive/mixed failure percentage, cohesive failure within composite was observed as

Table 2. Microtensile bond strength (mean \pm SD) for five self-etching adhesive systems after 24 h and 3 months in water. Values having different capital letters in lines (comparison of the five dentin bonding agents within the same storage time) and different lower case letters in a column (comparison of the two storage times for same dentin bonding agent) exhibited statistically significant differences ($p < 0.05$).

| Water storage time | <i>n</i> | Optibond All-In-One | <i>n</i> | G Aenial Bond | <i>n</i> | Clearfil SE Bond | <i>n</i> | Adper Prompt L Pop | <i>n</i> | Futurabond M |
|--------------------|----------|---------------------|----------|-------------------|----------|-------------------|----------|--------------------|----------|-------------------|
| 24 h | 61 | 30.4 ± 5.3 Aa | 64 | 35.3 ± 6.2 Ba | 57 | 43.9 ± 6.8 Ca | 59 | 26.2 ± 6.2 Da | 58 | 18.4 ± 3.8 Ea |
| 3 months | 72 | 28.1 ± 5.6 Aa | 60 | 32.7 ± 5.9 Ba | 57 | 42.1 ± 6.6 Ca | 58 | 21.2 ± 5.8 Db | 51 | 17.0 ± 3.5 Da |

Table 3. Distributions of failure patterns for each experimental condition: adhesive/mix, cohesive resin, cohesive dentin).

| Dentin bonding agent | Water storage time | Adhesive/mix | Cohesive resin | Cohesive dentin |
|----------------------|--------------------|--------------|----------------|-----------------|
| Optibond All-In-One | 24 h | 93.5% | 4.9% | 1.6% |
| | 3 months | 91.68% | 6.94% | 1.38% |
| G Aenial Bond | 24 h | 92.3% | 6.2% | 1.5% |
| | 3 months | 85% | 10% | 5% |
| SE Bond | 24 h | 87.79% | 7.01% | 5.2% |
| | 3 months | 89.49% | 7.01% | 3.5% |
| L Pop | 24 h | 94.92% | 5.08% | – |
| | 3 months | 100% | – | – |
| Futurabond M | 24 h | 100% | – | – |
| | 3 months | 100% | – | – |

the second most common failure mode for all except Futurabond M.

Consistent with results of this study, similar bond strength values after 24 h were reported in other studies for Clearfil SE Bond.[15,16] Proença et al. [15] found that Clearfil SE Bond exhibited the highest μ TBS value compared with L Pop and Futurabond M. The superior performance of this system is attributed to the synergy of an unsaturated 10-MDP (10-Methacryloyloxydecyl dihydrogen phosphate) as the acidic monomer combined with HEMA (2-hydroxyethyl methacrylate), which supposedly improves tooth surface wetting and calcium ion chelation in dentin.[15,16]

Van Landuyt et al. [17] found that μ TBS was significantly poorer with Optibond AiO than with Clearfil SE Bond and G Bond. Optibond AiO incorporates not only HEMA (8%–11%) but also water, acetone (35%–45%)

and ethanol (4%–9%). This can result in stronger water absorption by the adhesive resin, which could act as a semi-permeable membrane allowing water transport and thus affect the adhesive's mechanical properties.[18] Another possible explanation for the relatively lower performance of Optibond AiO is water entrapment within the hybrid layer [19] because of high hydrophilicity; incomplete solvent removal from the adhesive layer [5] can also adversely affect Bi-GMA polymerization [20] and decrease the bond strength of Optibond AiO.

G Aenial Bond exhibited the highest μ TBS values after Clearfil SE Bond, probably because of 4-MET in the former. This monomer can chemically interact with hydroxyapatite crystals.[21] G Aenial Bond is HEMA-free. HEMA promotes adhesion in most self-etch systems, [22] maintains resin monomers in one solution, and prevents phase separation.[23,24] However, high HEMA

Table 4. Distribution of microleakage scores, the mean values and standard deviations of enamel and dentin microleakage for each group of studied dentin bonding agents and pair wise comparisons. Within a column, values having different capital letters exhibited statistically significant differences ($p < 0.05$); comparison of the dentin bonding agents within each groups. Within a row having different lower case letters exhibited statistically significant difference ($p < 0.05$); comparison of the same dentin bonding agent between enamel and dentin within each group).

| Dentin bonding agents | n | Enamel leakage scores | | | | | | | | Dentin leakage scores | | | | | | | | |
|-----------------------|----|-----------------------|----|---|---|---|------|-----------|-----------|-----------------------|----|----|---|----|---|------|-----------|-----------|
| | | 0 | 1 | 2 | 3 | 4 | Mean | Std. dev. | Dunn test | n | 0 | 1 | 2 | 3 | 4 | Mean | Std. dev. | Dunn test |
| 24 h | | | | | | | | | | | | | | | | | | |
| Futurabond M | 42 | 25 | 17 | – | – | – | 0.4 | 0.49 | Aa | 42 | 11 | 25 | 6 | – | – | 0.88 | 0.63 | Ab |
| Clearfil SE Bond | 42 | 39 | 3 | – | – | – | 0.07 | 0.26 | Ba | 42 | 35 | 7 | – | – | – | 0.17 | 0.37 | BCb |
| Optibond All-In-One | 42 | 18 | 24 | – | – | – | 0.57 | 0.5 | Aa | 42 | 37 | 5 | – | – | – | 0.12 | 0.32 | Cb |
| Prompt L Pop | 42 | – | 42 | – | – | – | 1 | 0 | Ca | 42 | 11 | 31 | – | – | – | 0.74 | 0.44 | Ab |
| G Aenial Bond | 42 | 22 | 20 | – | – | – | 0.48 | 0.5 | Aa | 42 | 25 | 17 | – | – | – | 0.4 | 0.49 | Ba |
| 3 months | | | | | | | | | | | | | | | | | | |
| Futurabond M | 42 | 24 | 18 | – | – | – | 0.43 | 0.5 | Aa | 42 | – | 17 | 8 | 17 | – | 2 | 0.91 | Ab |
| Clearfil SE Bond | 42 | 39 | 3 | – | – | – | 0.07 | 0.26 | Ba | 42 | 35 | 7 | – | – | – | 0.17 | 0.37 | Bb |
| Optibond All-In-One | 42 | 18 | 24 | – | – | – | 0.57 | 0.5 | ACa | 42 | 20 | 17 | 5 | – | – | 0.64 | 0.69 | Ca |
| Prompt L Pop | 42 | – | 42 | – | – | – | 1 | 0 | Da | 42 | 2 | 38 | 2 | – | – | 1 | 0.31 | Da |
| G Aenial Bond | 42 | 9 | 33 | – | – | – | 0.79 | 0.41 | CDa | 42 | 8 | 34 | – | – | – | 0.81 | 0.39 | CDa |

Table 5. Comparison of the same dentin bonding agents between two groups (24 h and 3 months). Within a column, values having different capital letters exhibited statistically significant difference for enamel and dentin margins, separately ($p < 0.05$); comparison of the same dentin bonding agents between two groups.

| | Group | Futurabond M | Clearfil SE Bond | Optibond AIO | Prompt L Pop | G Aenial Bond |
|--------|-------|--------------|------------------|--------------|--------------|---------------|
| Enamel | 1 | A | A | A | A | A |
| | 2 | A | A | A | A | B |
| Dentin | 1 | A | A | A | A | A |
| | 2 | C | A | B | A | B |

concentrations can adversely impact the mechanical properties of the polymer over time. Higher HEMA concentrations in one-step self-etch adhesives may decrease the initial bond strength because of water attraction.[23] Therefore, the success of G Aenial Bond can be attributed to not only 4-MET presence but also HEMA absence.

Futurabond M is a one-step self-etching adhesive containing HEMA and acetone, while it does not contain water, which is required to dissociate weak acids into ionized forms for smear layer permeation and underlying intact tooth substrate demineralization. This probably affects the μ TBS values of Futurabond M. Furthermore, residual water in dentin channels and acetone should be completely eliminated before polymerization.[24] However, the recommended application time may be insufficient to allow water removal from the polymerized layer, [20] possibly resulting in the low μ TBS values for Futurabond M in this study. Another possible explanation is the increased initial acetone content that results in thinner adhesive layers, affecting the adjacent dentin bond.[25]

The bond strengths after 24 h decreased for all adhesives during 3 months of water storage, with the decrease being insignificant only except L Pop. Water storage for 3 and 6 months did not significantly decrease the μ TBS values of G Bond and Clearfil S3 Bond,[26] consistent with this study. Additionally, Osorio et al. showed that Clearfil SE Bond exhibited the best resin–dentin bond durability compared with Futurabond M and L Pop.[27] Clearfil SE Bond can produce hybrid layers that are less sensitive to age. The high camphorquinone percentage in this adhesive may have improved the polymerization degree [28] and indirectly improved the resistance to deterioration.

Moreover, G Aenial Bond showed better resistance to water degradation compared with L Pop after 3 months of water storage. Torkabadi et al. [26] evaluated the bonding durability of G Bond (HEMA-free) and Clearfil S3 Bond (HEMA-containing) after 1 year of water storage. There were no significant differences for G Bond between each time point, although μ TBS values decreased over time. Considering the deteriorating effects of HEMA on the adhesive interface over time, HEMA removal can increase the resistance of adhesives to deterioration.

Three month water storage significantly decreased μ TBS values for L Pop. Reis et al. [29] reported a

significant decrease in the bond strength of L Pop after 6 months of water storage (24.3 ± 3.1 MPa to 16.9 ± 4.1 MPa). Previous studies have shown that aggressive one-step self-etching adhesives (L Pop, Futurabond M) can completely dissolve the smear layer and form relatively thick hybridized complexes that incorporate the dissolved smear layer components.[30] Adhesive systems containing methacrylated phosphoric acid HEMA-ester (L Pop) may be quite acidic, resulting in profound enamel and dentin demineralization with the added disadvantage of hydrolytic instability.[31] These factors may explain the significant decrease in μ TBS values of L Pop after 3 months.

Microleakage

Data for distributions and mean values of enamel and gingival margin microleakage in each group and pair-wise comparisons are shown in Table 4. After 24 h, enamel margin microleakage was significantly less in the Clearfil SE Bond-treated teeth than in the other teeth. L Pop showed the highest microleakage score among all adhesives. Gingival margin microleakage was significantly lesser with Optibond AiO and Clearfil SE Bond than with the other bonding agents. Futurabond M showed the highest microleakage score among all adhesives. Enamel and gingival margin microleakage scores were significantly different for all adhesives except G Aenial Bond at 24 h.

After 3 months, enamel margin microleakage was significantly lesser in Clearfil SE Bond-treated teeth than in the other teeth. L Pop showed the highest microleakage score among all adhesives (Table 5). Gingival margin microleakage was significantly lesser in Clearfil SE Bond-treated teeth than in the other teeth. Futurabond M showed the highest microleakage score among all adhesives. Optibond AiO exhibited significantly lesser microleakage than L Pop. Enamel and gingival margin microleakage scores were significantly different for Clearfil SE Bond and Futurabond M.

Finally, enamel margin microleakage was significantly different between 24 h and 3 months with G Aenial Bond, while gingival margin microleakage was significantly different between time points with Futurabond M, Optibond AiO and G Aenial Bond (Table 5).

Clearfil SE Bond displayed the lowest microleakage scores in this study. Deliperi et al. [32] indicated that Clearfil SE Bond showed low microleakage values, similar to those of the total etch system Prime & Bond NT. Osorio et al. [33] reported the lowest dye penetration values in dentin for Clearfil SE Bond compared with Etch & Prime 3.0 and Scotchbond MP. Condon et al. [34] reported that apart from 10-MDP, silanated colloidal silica nanofillers in Clearfil SE Bond increase crosslinking and decrease polymerization shrinkage, which may contribute to decreased microleakage.

L Pop exhibited relatively high leakage scores compared with the other adhesives at both storage times. Gueders et al. [35] reported significantly high microleakage scores for L Pop than for Scotchbond MP, Optibond Solo Plus and i Bond. Similarly, Manuja et al. [36] reported that L Pop showed higher microleakage scores than Optibond AiO. According to Brackett et al. [37], the pH of the adhesive is not the only factor determining sealing ability. Other factors such as substrate-related factors, variations in adhesive viscosity, surface tension, functional monomers, water concentration and other components of the bonding system also affect bonding.[36,38] In contrast, Van Landuyt et al. [17] and Blunck and Zaslansky [39] demonstrated no correlation between pH and marginal integrity and gap formation in different adhesives.

Futurabond M exhibited the maximum gingival margin microleakage and very little enamel margin microleakage. Amaral et al. reported that the presence of acetone results in high microleakage scores in a moist environment.[7] Systems with only acetone and no water are more sensitive to moisture variations. The advantage of non-acetone adhesive systems is relative insensitivity to dentin surface moisture.[40] This can be the reason for the high gingival margin microleakage scores for Futurabond M.

Blunck and Zaslansky [41] reported that all-in-one adhesives exhibited a decreased marginal quality score and a varied deterioration rate; furthermore, different materials had different deterioration rates after short- and long-term water storage (21 days, 1 year, 3 years). Monticelli et al. [42] reported a decreased sealing effectiveness of Clearfil SE Bond and L Pop along enamel and dentin margins after 24 months of water storage. Researchers report that water sorption phenomena may occur over time, resulting in swelling of the resin layer and weakening of the adhesive joint.[38,42] Phase separation and/or the low polymerization rate of simplified adhesives reportedly result in the formation of weaker interfaces with exposed collagen fibrils, which possibly degrade over time.[43]

Taken together, water and other chemicals leaching from the oral cavity may decrease the mechanical properties of polymers.[44] In addition, the substrate is a biological tissue, which makes adhesion difficult.[45] Additionally, the chemical composition of the adhesive

itself plays an important role in forming a strong, durable and biologically compatible bond.[46]

Conclusion

Our first null hypothesis was accepted because the results of this study confirm that the water-free, acetone-based, HEMA-containing all-in-one adhesive Futurabond M exhibited the lowest μ TBS values not only at 24 h but also after 3 months of water storage. Also, the microleakage scores showed that Futurabond M is very sensitive to dentin moisture, as indicated by the higher gingival margin microleakage scores. Furthermore, ethanol–water-based adhesives used in this study displayed higher μ TBS values and lower leakage scores. The second null hypothesis can be accepted because, in any case, the μ TBS values of one-step self-etch adhesives were significantly lower than those of two-step self-etch adhesives; however, the μ TBS values of all-in-one adhesives differ according to the adhesive composition (more than the adhesive mechanism) and the μ TBS values of one-step self-etch adhesives are rather different. The same result is valid for the microleakage test. The third hypothesis can be accepted because the bond strengths of all adhesives at 24 h decreased during 3 months of water storage. The sealing effect of the adhesives at the enamel and gingival margins varied after 24 h depending on the adhesive type. Also, water storage and thermocycling slightly increased the microleakage scores over time, and this increase was pronounced at the gingival margins after 3 months.

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