

Evaluation of Microtensile Bond Strength, Microleakage Pattern and Micromorphological Analysis of New Functional Monomer-Based Universal Adhesive Tooth Interface

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Abstract:

Objective: This study was intended to evaluate the micro tensile bond strength (μ TBS), micro leakage pattern, and micro morphological analysis of new 10-MDP-based universal adhesive/tooth interface and compare it with other two 10-MDP-based adhesive systems before and after storage. **Materials and Methods:** Two Universal adhesives (Single Bond Universal Adhesive and Ambar Universal) and one all-in-one adhesive (CLEARFIL S3BOND) were used. A total of 102 permanent human molars were used in this study. The teeth were randomly divided into three groups (n=34) according to the type of used adhesive; Ambar Universal (AU), CLEARFIL S3BOND (CTS), and Single Bond Universal (SBU). Each group was then subdivided into two subgroups (n=17) according to the time of testing. **Results:** It was shown that changing time had no statistically significant effect on μ TBS ($p > 0.05$); however, changing group had a statistically significant effect ($p < 0.05$). Additionally, the combination of changes in group and time had no statistically significant effect on the μ TBS ($p > 0.05$). Furthermore, it was shown that changing group or time did not significantly affect the micro leakage score. Finally, a slight difference was observed between the studied groups mainly in the resin tags; however, all adhesives showed a loss of resin tags after thermo cycling. **Conclusions:** The new 10-MDP-based universal adhesive had lower bond strength to dentin than the other tested adhesives. However, the new 10-MDP-based universal adhesive/tooth interface had relatively similar micro leakage patterns and micro morphological features in comparison with the other tested adhesives.

Introduction:

Adhesive systems action based on a dual adhesion principle, where one side binds to the dental substrates while the other binds to the composite restoration. The latter occurs by a co-polymerization process, while the former occurs assumedly by micromechanical adhesion.¹ Micromechanical adhesion is accomplished by an exchange action in which minerals (mainly hydroxyapatite) demineralized from the dental substrates by acids are replaced by resin monomers which are then polymerized and interlocked in the porosities providing micromechanical retention.^{2, 3} Furthermore, incorporating certain monomers that have an affinity for hydroxyapatite (HAp) can create chemical adhesion between the adhesive system and the dental substrates.⁴ The main factors in obtaining a successful restoration are marginal sealing and bond durability to the dental substrates.⁵ However, the tooth structure comprises two different hard tissues enamel and dentin. It is well-known that achieving a similar bonding capacity to both tissues is challenging because dentin is more hydrophilic and contains more organic content than enamel which renders bonding to dentin more challenging.⁶

The improvement of adhesive systems over the last five decades has delivered several materials that can be used reliably in different restorative procedures. Currently, four types of adhesive systems are available in the market: Etch and Rinse adhesives (E&RAs) which depend on conditioning the dental substrates before bonding application, Self-Etch adhesives (SEAs) which rely on the adhesive itself to etch and infiltrate into the dental substrates without the need for prior conditioning, and resin-modified glass ionomer (RMGI) and universal adhesives which can be used in etch and rinse (E&R) or self-etch (SE) modes according to clinician preference.⁷ Despite the differences between adhesive types, their composition tends to be somewhat similar; however, the proportions of their ingredients vary from one system to another. Essentially, adhesives consist of resin monomers, solvents, initiators, inhibitors, and often fillers. Also, they could contain unique ingredients that offer distinctive functions to the adhesive. Understanding the behavior of each ingredient in these adhesives plays a major role in obtaining predictable and reliable clinical outcomes.⁵ Long-term restorations are what provide evidence for the success of an adhesive system. However, even with the continuous development of adhesive systems, the interface between the resin and dental substrates in composite restorations remains the weakest point. The majority of adhesive systems available today show outstanding immediate and short-term bonding capability, but their long-term durability remains doubtful. The failure rate of composite restorations can get to 15-20% after 12 years; this failure is caused mainly by wear, marginal defects, and

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secondary caries.^{8, 9} Consequently, the application of adhesive systems is considered technique-sensitive where a minor error in adhesive system application will result in early marginal degradation or quick debonding.² Therefore, current research is focused on the development of new adhesive systems to overcome these challenges. Furthermore, researchers attempt to improve current adhesive systems by adding certain chemicals to their composition or by modifying their bonding protocol to stabilize or even increase the bond strength between the resin restoration and dental substrates.¹⁰ Hence, this study was conducted to evaluate the μ TBS, micro leakage pattern, and micromorphological analysis of a new 10-MDP-based Universal adhesive and compare it with two other 10-MDP-based adhesive systems after 24h and after 5,000 cycles of thermocycling.

This study was also conducted to test the null hypotheses that there is no significant difference between the new 10-MDP-based universal adhesive and the other tested adhesive systems in regards to: I. μ TBS to dentin, II. occlusal and gingival microleakage patterns of the adhesive/dentin interface, and III. Micromorphological features of the adhesive/dentin interface

Materials and Methods:

Materials: Two different Universal adhesives (Single Bond Universal Adhesive and Ambar Universal) and one all-in-one adhesive (CLEARFIL S3BOND) were used in this study.

Methods: Teeth selection: A total of 102 permanent human molars were used in this study. The teeth were collected from patients seeking extraction treatment in the outpatient clinic oral surgery department, Faculty of Dentistry at Mansoura University, according to the regulation of our institution's ethical committee. Teeth were cleaned thoroughly from any calculus and soft tissue deposits using a hand scaler followed by a low-speed rubber cup with prophyl paste and then washed with distilled water. After that, teeth were examined using a stereomicroscope (SZ TP, Olympus, Tokyo, Japan) to exclude those with pre-existing defects, cracks, or restorations. The teeth were then stored in distilled water until use and water was changed every week. **Study Groups:** All teeth (n=102) were randomly divided into three groups (n=34) according to the type of used adhesive. Group one (AU) was bonded with Ambar Universal, the second group (CTS) was bonded with CLEARFIL S³BOND, and the third group (SBU) was bonded with Single Bond Universal. Each group was then subdivided into two subgroups (n=17) according to the time of testing (immediate/delayed). The immediate subgroups were tested immediately after 24h, while the Delayed subgroups were tested after thermo cycling (5,000 cycles). **Micro tensile bond strength (μ TBS) test:** For this test 7 teeth were tested from each subgroup with a total number of 42 teeth. The facial enamel of each tooth was removed

using a low-speed diamond disc (IsoMet 4000 saw, Buehler Ltd., Lake Bluff, USA) under sufficient water cooling to expose the facial dentin surface. The dentin surface was then polished with 600-grit silicon carbide paper under water flow to establish a fixed smear layer thickness. The dentin surface was dried with a micro-brush leaving it slightly moist and shiny. After that, all the adhesives were applied on the dentin surface in self-etch strategy and following the manufacturer instructions. The bonded surface was then covered by a nanohybrid bulk-fill resin composite restorative material (Tetric N-Ceram Bulk Fill, Ivoclar Vivadent). The composite was added in 2-3 increments to form a 3-4 mm composite block. Each increment was light-cured according to the manufacturer's instructions using an LED curing light (COXO, DB-686 DELI, LED Curing light) (1200mW/cm² with a wave length between 450-470nm). The intensity of LED curing light was monitored using a radiometer (Demetron LC, Kerr, Germany). The teeth were then embedded in acrylic resin blocks (Acrostone, Cairo, Egypt). The acrylic blocks were mounted in a diamond-cutting automated saw (Isomet 4000, Buehler Ltd., Lake Bluff, IL, USA), then the teeth were cut vertically and horizontally in a bucco-lingual direction under water coolant to create composite-adhesive-dentin beams with a surface area of 1 x 1 mm². All the beams of each tooth were tested with a maximum of 5 beams per tooth. The score for each tooth represents the mean μ TBS of the beams of that tooth. The beams were glued at their composite and dentin ends in Geraldeli's jig's middle groove with cyanoacrylate-based glue (Zapit, DVA Inc, USA). The beams were then mounted onto a universal testing machine (Instron model 3345, England) using Geraldeli's jig. The universal testing machine was used to apply a tensile load to the specimen with a cross-head speed of 0.5 mm/min until specimen fracture. The μ TBS scores were calculated in Mega Pascal (Bluehill Lite software, Instron model 3345, England). The mode of failure for each specimen was then determined using a stereomicroscope (Nikon MA 100, Tokyo, Japan) at x30 magnification. Additionally, scanning electron microscopy (SEM) (JSM-651 OLV, JEOL Japan) was used to observe the fracture planes of all failure modes except for the cohesive fracture in composite. **Micro leakage test:** For this test 7 teeth were tested from each subgroup with a total number of 42 teeth. A box-shaped class V cavity with occlusal bevel was prepared on the buccal surface of each tooth with a mesio-distal width of 3-4 mm, occluso-gingival height of almost 3 mm and a depth of 2-2.5 mm. The cavity was placed with the occlusal margin being on enamel and the gingival margin on dentin. The cavity was prepared using a diamond straight fissure point (Mani SF-41) in a high-speed handpiece cooled with an air-water spray, the point was changed after every 5 cavities.¹¹ Before bond application, the enamel was selectively etched by 37% phosphoric acid etching gel (Meta etchant, Meta Biomed Co. Ltd.) for 15 seconds, then rinsed with an air/water syringe and dried with a gentle air stream leaving the dentin slightly moist and shiny. After that,

all the adhesives were applied on the dentin surface in self-etch strategy and following the manufacturer's instructions. The cavity was then restored with composite in 2 increments, and each increment was light-cured according to the manufacturer's instructions. The restoration was then finished by composite finishing burs and polished by polishing discs. The teeth were then embedded in acrylic resin blocks (Acrostone, Cario, Egypt). All tooth surfaces were covered with nail varnish except for the class V composite restoration and 1 mm from its margins. The teeth were then immersed in 2% methylene blue dye for 24h at 37°C. After dye exposure, the teeth were rinsed thoroughly with water to ensure the removal of the dye. After that, each tooth was divided longitudinally in a bucco-lingual direction into two halves using a diamond-cutting automated saw (Isomet 4000, Buehler Ltd., Lake Bluff, IL, USA) under water coolant. Microleakage in both occlusal and gingival margins was examined using a stereomicroscope (Nikon MA 100, Tokyo, Japan) at x30 magnification. The results were evaluated based on the following scoring system¹¹:

Score (1) Negative dye penetration, Score (2) Dye penetration not surpassing the middle of the cavity Depth, Score (3) Dye penetration exceeds the middle of the cavity depth, and Score (4) Dye penetration extends over the axial wall.

Micromorphological analysis of the adhesive/dentin interface: For this test 3 teeth were tested from each subgroup with a total number of 18 teeth. A cavity was prepared and restored for each tooth similar to the microleakage test. Each tooth was divided longitudinally in a bucco-lingual direction into two halves using a diamond-cutting automated saw (Isomet 4000, Buehler Ltd., Lake Bluff, IL, USA) under water coolant. Only one half from each tooth was tested while the other half was discarded. Each tested half was polished with 600-, 1000-, 1200-, and 2000-grits silicon carbide paper under water. Final polish was achieved by using a polishing cloth with fine diamond pastes (6 um, 4 um, and 1 um) (MetadiTM, Buehler, Lake Bluff, IL, USA).¹² After that, all specimens were cleaned ultrasonically in a digital ultrasonic water bath for 10 minutes (Guilin Woodpecker, Guangxi, China). In order to create an acid-base challenge, the specimens were subjected to a 10% orthophosphoric acid solution or 5 seconds and then to a 5% sodium hypochlorite solution for 5 minutes.¹² The aim of subjecting the Specimens to an acid-base challenge is to demineralize any dentin that was not infiltrated by resin so that the

dentin could be desiccated. The specimens were then gold-sputtered (SPI Module-Sputter Carbon / Gold Coater, EDEN instruments, Japan) and viewed under a scanning electron microscope (JSM-6510, JEOL, Japan) in secondary electron detection mode at an accelerating voltage of 30 KV, working distance of 10-15 mm, at X500, X1000, and X2000 magnifications.

Statistical analysis:

The data were collected, tabulated and statistically analyzed using SPSS™ Software (V.22. IBM, NY, USA). Qualitative data were described using number and percentage. Quantitative data were described using mean for non-normally distributed data after testing normality using Shapiro-Wilk test. Significance of the obtained results was judged at the (p=0.05) level.

Results:

Microtensile bond strength test:

Shapiro-Wilk test showed that the μTBS data of all groups did not follow a normal distribution pattern (p > 0.05). Therefore, log transformation was done to use two-way ANOVA test on a non-normally distrusted data. Two-way ANOVA test showed the combined effect of changing group (AU, CTS, SBU) and time (immediate and delayed) on the μTBS (Table 1).

Table 1: Two-way ANOVA test used for determining the effect of changing group and time on the μTBS

Source	Type III Sum of Squares	df	Mean Square	F	P-Value
Corrected Model	875.2a	5	175.04	4.45	.003
Intercept	19001.73	1	19001.73	483.15	.000
Time	59.86	1	59.86	1.52	0.226
groups	684.18	2	342.09	8.7	0.001
Time * groups	105.79	2	52.9	1.35	0.274
Error	1376.51	35	39.33		
Total	21350.44	41			
Corrected Total	2251.71	40			
a. R Squared = .389 (Adjusted R Squared = .301)					

Table 2: Post-Hoc Tukey test used for pairwise comparison in μTBS between the groups with disregard to time

	AU	CTS	SBU
Mean±SD (Immediate and Delayed)	17.13±5.37 ^a	20.16±6.32 ^b	27.12±7.18 ^{ab}

Similar superscripted letters denote significant difference between groups within same row.

Table 3: Comparison of occlusal and gingival margins micro leakage scoring between studied groups in immediate and delayed, and between immediate and delayed in each group

Occlusal margin		AU	CTS	SBU	p-Value
Immediate	Score 1	6 (85.7%)	6 (85.7%)	7 (100%)	P=0.392
	Score 2	0	0	0	
	Score 3	0	1 (14.3%)	0	
	Score 4	1 (14.3%)	0	0	
Delayed	Score 1	7 (100%)	7 (100%)	7 (100%)	P=1.0
	Score 2	0	0	0	
	Score 3	0	0	0	
	Score 4	0	0	0	
SM test		P=0.317	P=0.317	P=1.0	
Gingival margin		AU	CTS	SBU	p-Value
Immediate	Score 1	4 (57.1%)	5 (71.4%)	4 (57.1%)	P=0.968
	Score 2	2 (28.6%)	1 (14.3%)	2 (28.6%)	
	Score 3	0	0	0	
	Score 4	1 (14.3%)	1 (14.3%)	1 (14.3%)	
Delayed	Score 1	2 (28.6%)	4 (57.1%)	4 (57.1%)	P=0.684
	Score 2	3 (42.9%)	1 (14.3%)	3 (42.9%)	
	Score 3	1 (14.3%)	1 (14.3%)	0	
	Score 4	1 (14.3%)	1 (14.3%)	0	
SM test		P=0.317	P=0.414	P=0.414	

The test indicated that changing time had no statistically significant effect on μ TBS ($p>0.05$); however, changing group had statistically significant effect ($p<0.05$). Additionally, the test showed that the combination of changes in group and time had no statistically significant effect on the μ TBS ($p>0.05$) with 41.8 % of μ TBS change was affected by changing group only. The Post-Hoc Tukey test was used for pairwise comparison in μ TBS between the groups with disregard to time (Table 2). This test showed that there was a statistically significant difference in the μ TBS between SBU and the other two groups (AU and CTS) ($p<0.05$). However, there was no statistically significant difference in the μ TBS between AU and CTS ($p>0.05$). AU showed premature failure of one tooth in the delayed group, while CTS and SBU did not show premature failure. SBU showed the highest immediate and delayed μ TBS, followed by CTS then AU. The predominant failure type was an adhesive failure in groups AU and CTS in immediate and delayed, and cohesive failure in composite in group SBU in immediate and delayed. **Microleakage test:** The immediate and delayed micro leakage scores and percentages of the occlusal and gingival margin are shown in table 3. Monte Carlo test showed no significant difference between the three studied adhesive materials both in occlusal and gingival margin in immediate and delayed results ($p>0.05$). Additionally, the Stewart Maxwell test revealed that there is no significant difference in the micro leakage scores between the immediate and delayed subgroups among all adhesives in both occlusal and gingival margins ($p>0.05$). The microleakage scoring comparison of immediate and delayed of occlusal and gingival margins between studied groups are shown in Table 3.

Micro morphological analysis of the adhesive/dentin interface: All the tested adhesives examined before storage showed a uniform thin hybrid layer and cylindrical-shaped resin tags. The resin tags of the tested adhesives had similar thickness and shape; however, they were different in length and number. AU had numerous short resin tags, while SBU had numerous long resin tags. CTS had long resin tags but less in number when compared with AU and SBU. On the other hand, no resin tags were present after storage for all the tested adhesives; however, there was no disintegration in the hybrid layer.

Discussion:

Universal adhesives rely mainly on their functional monomer and its chemical bonding capability to achieve higher bond strength and durability. The most commonly used functional monomer in universal adhesives is 10-MDP due to its superior performance over other functional monomers.¹³⁻¹⁵ However, 10-MDP-based universal adhesives showed variable bonding results despite having the same functional monomer. This is attributed mainly to the different concentrations and purities of 10-MDP in universal adhesives.¹⁶ Moreover, the concentration of other components in the adhesive system can affect the reactivity and chemical bonding potential of 10-MDP.¹⁷ Regarding the μ TBS test, despite having the same functional monomer, the adhesive systems in the present study had different dentin bonding strength values when compared with each other. This shows that the adhesives' bonding ability is material-dependent, disregarding the similarity in the functional monomer. This indicates that the compositions of the

adhesive systems in this study, including type and quantity of functional monomers, hydrophilic and hydrophobic components, solvents, photo initiator system, and other factors, all play a role in the bonding performance.¹⁸ In fact, Fujita et al.¹⁷ stated that the rate of 10-MDP Ca-salts formation is dependent on the components that the adhesive comprises more than the quantity of the functional monomer itself. On the contrary, the dentin bonding performance of the adhesives used in this study was not significantly affected by the storage time. This is mainly due to the presence of 10-MDP functional monomer in the adhesives' composition, which forms strong and stable chemical bonds with HAp.¹³ A study by Inoue et al.¹⁹ found that the μ TBS of an adhesive system containing 10-MDP (Clearfil SE) remained stable even after 100,000 cycles. The bond stability of 10-MDP-based adhesive systems is attributed to the formation of stable 10-MDP Ca-salts and to the nano-layering between the 10-MDP and HAp.²⁰ The results showed that the immediate μ TBS of SBU was significantly higher than both AU and CTS.

This might be attributed to the presence of polyalkenoic acid copolymer (PAC), which also bonds chemically to HAp.²¹ Conversely, Munoz et al.²² stated that PAC had adverse effects on the bond strength of 10-MDP-based adhesives, as it competes with 10-MDP on binding sites. On the other hand, AU had the lowest initial bond strength. A presumption was made that the APS (advanced polymerization system) photo initiator system of the adhesive was the reason behind the low bond strength. However, a study by Basilio et al.,²³ found that the APS photo initiator system did not affect the bond strength of the adhesive. Therefore, further studies are required to evaluate the dentin bonding ability of this adhesive.

Regarding the failure modes, AU showed the highest AD failure among the tested adhesive systems. This is mainly due to the double layer application instructed by the manufacturer, which increased the thickness of the adhesive layer and made it susceptible to fracture. Similar to the bond strength, the failure mode of all tested adhesive systems did not change after thermo cycling. In addition, there was a negative correlation between the bond strength of the adhesive and the amount of AD failure. This correlation was also present in the studies of Moll et al.²⁴ and Hamama et al.²⁵ Regarding the micro leakage test, there was no significant difference between the tested adhesive systems. This may be attributed to the selective enamel etching (SEE) strategy, which was used in this study for all the tested adhesive systems. The SEE strategy combines the E&R strategy on enamel with the SE strategy on dentin. It has been proven that etching enamel before applying mild or ultra-mild adhesive systems is recommended to ensure better bonding performance on enamel.²⁶ On the other hand, utilizing the SE strategy on dentin theoretically ensures complete adhesive infiltration into the dentin substrate

due to the simultaneous demineralization and infiltration.²⁷ Additionally, the storage time did not affect the micro leakage scores of all tested groups. This can also be attributed to the application strategy used in this study, as well as the presence of the 10-MDP functional monomer, which stabilized the bonding performance of the tested adhesives. Furthermore, the findings of this study showed that there is no relation between the μ TBS and the micro leakage scores, which was in agreement with Cenci et al.²⁸ Moreover, there was no significant difference between the occlusal and gingival micro leakage scores before and after storage except for AU, which showed a significant difference after storage. However, numerically, all tested adhesives showed lower micro leakage scores in the gingival margin before and after storage. These findings support the fact that adhesion to enamel is stronger than adhesion to dentin, especially in the SEE strategy.²⁶ Regarding the micro-morphological analysis, all tested adhesives showed relatively similar results with slight difference in the length and number of resin tags in the specimens tested before storage. The absence of resin tags in the specimens tested after storage suggests that resin tags are not responsible for the adhesive's bond strength, since the μ TBS of the tested adhesives did not change after storage. Therefore, there is no relation between the absence or presence of resin tags and the bond strength of the adhesive, which was also confirmed by other studies.^{29,30} This also emphasizes the importance of the quality of the hybrid layer on the bond performance, especially when the adhesive is applied in the SE strategy.^{26,31}

The findings of the micro-morphological analysis in this study were in disagreement with the study done by Inoue et al.,(2005)¹⁹ in which the micromorphology of the adhesive/dentin interface of a 2-SEA containing 10-MDP (Clearfil SE) did not change even after 100,000 cycles. This may be attributed to the difference in the adhesive system used in each study. Fundamentally, 2-SEAs involve the separate application of an acidic functional monomer (10-MDP) before applying the adhesive, which not only improves the action of the functional monomer but also provides a better seal by a hydrophobic adhesive layer.³² Furthermore, De Munck et al.,(2005)³³ stated that the micro-morphological features of the adhesive/dentin interface of SEAs are greatly dependent on the level and kind of interaction between the functional monomer and the dental substrate.

In light of the results of this study, the first null hypothesis, which states that there is no significant difference between the new universal adhesive and the other tested adhesive systems in μ TBS, was rejected. While the second and third null hypotheses, which state that there is no significant difference between the new universal adhesive and the other tested adhesive systems in micro leakage pattern and micro-morphological features, were accepted.

Conclusions:

Based on the results of this study taking into consideration its limitations, the following conclusions were made:

1. The new 10-MDP-based universal adhesive showed lower bond strength to dentin than the other tested adhesives.
2. The adhesive/dentin interface of all tested adhesives had relatively similar micro leakage patterns and micro morphological features.
3. The dentin bond strength did not affect the microleakage patterns and micromorphological features of the adhesive/dentin interface.
4. Aging did not affect the dentin bond strength of the adhesives used in this study.
5. Resin tags did not impact the dentin bond strength of the adhesives used in this study.

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