BOND STRENGTH AND INTERFACIAL MORPHOLOGY OF A MULTI-MODE ADHESIVE RESIN CEMENT TO ENAMEL AND DENTIN

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Abstract:

Introduction: Resin cements are used for cementation of indirect restorations. A high-quality adhesion to tooth structure is primordial for the success of indirect cemented restorations.

Objectives: Were to assess the shear bond strength of RelyX ultimate adhesive resin cement to both enamel and dentin when used in different application modes (self-etch and total-etch) and to study resin-tooth interfaces using SEM.

Materials and methods: 40 enamel specimens and 40 dentin specimens were used in this study. Each substrate specimens were randomly divided into four groups of 10 specimens each according to resin cement applied: Enamel groups: Group A1: RelyX ultimate as total-etch; Group A2: RelyX ultimate as self-etch; Group A3: Variolink II; Group A4: Multilink automix. Dentin groups: Group B1: RelyX ultimate as total-etch; Group B2: RelyX ultimate as self-etch; Group B3: Variolink II; Group B4: Multilink automix. Resin cements were built over the surfaces following manufacturer's instructions in a plastic tube. All specimens were thermocycled (500 cycles, 5°/55°C, 15 sec dwell time) then shear bond strength was measured in MPa and data were analyzed using F-test (ANOVA) and Post Hoc test. Three specimens from each group were further sectioned, gold sputtered and evaluated under SEM.

Results: All enamel groups showed a statistically significant difference for shear bond strength with P <0.001. Group A1 revealed the highest mean value of 35.60 MPa. While for dentin, there was no statistically significant differences among groups with P=0.053. Interfacial morphology of RelyX ultimate in total-etch mode showed better interface morphology and improved tag length compared to respective self-etch counterpart

Conclusions: An etching step prior to multi-mode adhesive resin cement increased its bond strength values and improved its penetration pattern to both enamel and dentin, also length of resin tags formed by total-etch adhesives seems not to play an important role in determining bond strength

Keywords: *Resin cements, Adhesives, Multi-mode, dental substrates, bonding, interface*.

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INTRODUCTION

Tooth colored inlays, onlays, veneers and crowns are now routinely bonded to the tooth substrate via the use of adhesive resin cements as they have the ability to bond to both tooth structure and restoration. The integration reinforces both structures and reduces microleakage at the restoration-tooth interface, postoperative sensitivity, marginal staining and recurrent caries (1). A durable bond at the tooth-restoration interface is fundamental for long-term success of an adhesive restoration (2). An extremely relevant factor for the success of cementation is the type of adhesive used together with the cement because the former is responsible for the bonding between the tooth structure and the cement itself (3).

Traditional resin cements can be divided into two groups according to the adhesive system used to prepare the tooth prior to cementation. One group utilizes etch-and-rinse adhesive systems (two -or three-step), whereas for the other group, dental substrates are prepared using self-etch primers/adhesives (one- and two-step) (4).

The total-etch strategy requires previous acid etching of the enamel and dentin surfaces which leads to the formation of pores and permits mechanical interlocking to the enamel, and exposition of collagen fiber network of dentin, resulting in the formation of hybrid layer. However, this technique consists of several clinical steps that have to be rigorously followed, a fact that makes this method complex and time consuming (5). In addition, the possibility of over etching the dentin and the difficulty in clinically determining the ideal moisture point of the etched substrate may result in the presence of dentinal collagen exposed by the acid but not impregnated by the adhesive, affecting bond durability (6).

The self-etch strategy depends on acid monomers that etch the substrate and simultaneously fill the pores formed, so they reduce the working time and speed up the cementation process of indirect restorations. In addition the mild etching provided by these agents might be more favorable since it does not completely remove the smear plugs present in the tubules, thus preventing an increase in dentinal permeability and reducing the chance of postoperative sensitivity (7). However, there are conflicting reports regarding the enamel bond strengths of indirect restorations using self-etch cements as the acidity of non-rinsed acidic primers may negatively affect the bond strength to enamel (8).

The use of phosphoric acid before the application of non-rinsed adhesives or self-adhesive cements has been suggested to improve adhesion. The efficacy of enamel acid pretreatment has been questioned (9). also the effect of intentionally etching dentin prior to the application of self-etch adhesives has been studied with controversial results (10, 11).

Considering the differences in professional judgment regarding the selection of the adhesive strategy and number of steps, some manufacturers have released more versatile adhesive systems that include etch-and-rinse and self-etch options. These new materials are called universal or multi-mode adhesives (12-14).

Combining a universal multi-mode adhesive with a new resin cement gives it the versatility to cover all techniques and all indications as the cement can be used either in a self-etch mode or in a total-etch mode (15). The aim of the present in vitro study was to determine the performance of a new multi-mode adhesive resin cement when bonded to enamel and dentin in the different application modes and to assess the morphological characterization of the tooth/adhesive interface microscopically. The null hypothesis advanced in the study was that prior etching of enamel and dentin with phosphoric acid does not influence the bonding efficiency of multi-mode adhesive system.

MATERIALS AND METHODS

Forty extracted human permanent anterior teeth for shear bond strength of resin cements to enamel and 40 extracted human permanent molar teeth for shear bond strength of resin cements to dentin were used in this study. Selected teeth were free of caries, attrition, abrasion, cracking or previous restoration. The teeth were thoroughly washed with tap water, debrided from all soft tissues or bone, and stored in physiological saline at room temperature until use. Three resin cements with their respective adhesives were used in this study: RelyX ultimate (3MESPE, St.Paul, MN, USA), Variolink II and Multilink automix (Ivoclar Vivadent, Schaan, Liechtenstein) as shown in (Table 1).

Anterior teeth had their labial surface ground flat under running water using a diamond stone mounted to a high speed handpiece. Molar teeth had their occlusal surface ground flat by means of a mechanical grinder to remove enamel and expose flat dentinal surfaces, then the flattened surfaces were polished using #600 grit silicon carbide paper under running water. Roots were cut just cervical to the cemento-enamel junction with a diamond disc mounted on a low speed handpiece

Split-copper metallic molds of 20 mm length and 14 mm diameter were filled with auto polymerizing acrylic resin. The enamel specimens were embedded into the acrylic resin with their labial surface facing upwards, while the dentin specimens were embedded with their occlusal surface facing upwards. Each substrate specimens were then divided into four groups of ten specimens eachaccording to resin cement applied

Enamel groups: Group A1: RelyX ultimate in total-etch mode; Group A2: RelyX ultimate in self-etch mode; Group A3: Variolink II (total-etch); Group A4: Multilink automix (self-etch).

Dentin groups: Group B1:RelyX ultimate in total-etch mode; Group B2:RelyX ultimate in self-etch mode; Group B3:Variolink II (total-etch); Group B4: Multilink automix (self-etch). A thin walled plastic tube (3.5 mm diameter and 2 mm height) was carefully centered on the flat tooth surface and secured in position with sticky wax.The resin cements were applied inside the tube following strictly the manufacturer instructions (Table 1).

Enamel and dentinspecimenswere stored in distilled water at 37 ºC for 24 h and were then thermocycled 500 cycles between 5 ºC and 55 ºC with a dwell time of 15 seconds at each temperature (16). All specimens were then subjected to shear bond strength test using a universal testing machine

Table (1): List of material composition and application procedures

Material	Composition	Application procedures
RelyX Ultimate resin cement	**Base Paste:** Methacrylate monomers, Radiopaque silanated fillers, Initiator components, Stabilizers, Rheological additives **Catalyst Paste:** Methacrylate monomers, Radiopaque alkaline (basic) fillers, Initiator components, Stabilizers, Pigments Rheological additives, Fluorescence dye, Dark cure activator for Scotch bond Universal adhesive.	**Self-etch mode:** 1. Apply Scotchbond universal adhesive with brush and rub for 20 s, gently air dry for 5 s and light-cure for 10 s. 2. Dispense cement from the automix double-push syringe and light cure for 20 s. **Total-etch mode:** 1. Apply Scotchbond Etchant for 15 s, rinse with water for 10 s and then dry with cotton pellets. 2. Apply Scotchbond adhesive with brush and rub for 20 s, gently air dry for 5 s and light-cure for 10 s 3. Dispense cement from the automix double-push syringe and light cure for 20 s.
scotchbond Universal Adhesive	MDP Phosphate Monomer, Dimethacrylate resins, HEMA, polyalkenoic acid Copolymer, Filler, Ethanol, Water, Initiators, Silane.	
Scotch bond Universal Etchant	Phosphoric acid (35%), with a pH of approximately 0.1 colloidal silica thickener, color, water	
Variolink II resin cement	BisGMA, UDMA, TEGDMA, barium glass fillers, ytterbium trifluoride, boroalumino fluorosilicate glass, spheroidal mixed oxide, catalysts, stabilizers, pigments.	1. Apply total-etch phosphoric acid (37%) for 30 s on enamel and 15 s on dentin, rinse for 10 s and blot-dry with cotton pellet. 2. Apply Excite DSC with a brush and agitate 10 s, dry for 1–3 s and then light-cure for 20 s. 3.Dispense cement catalyst and base on a mixing pad and mix in a 1:1 ratio and apply into the plastic tube and light cure for 20 s.
Excite Dsc	DMA, HEMA, alcohol, phosphoric acid acrylate, SiO2, initiators, stabilizers	
Etchant gel	37%phosphoric acid in water , thickening agent and colorants.	
Multilink automix resin cement	**Multilink Base and Catalyst:** Bis-EMA, UDMA, bis-GMA, HEMA, barium glass filler, silicon dioxide filler, ytterbium trifluoride, catalysts, stabilizers, pigments	1. Mix Multilink Primers A and B in 1:1 ratio. 2. Scrub mixed primer on surface with brush for 30 s on enamel and 15 s on dentin. 3. Applied primer is subsequently dried with water- and oil-free air. 4. Dispense Multilink cement from automix double-push syringe then light cure for 20 s.
Multilink Primer A / B	**Multilink Primer A** is an aqueous solution of initiators. **Multilink Primer B** contains HEMA, phosphonic acid and methacrylate monomers	

(Comten Industries, INC., Florida, USA) with the stainless steel knife perpendicular to the junction between the tooth surface and the resin cement at a cross head 0.5mm/minute until failure occurred. The fracture load was recorded and the shear bond strength was calculated in MPa. Comparison of mean values of shear bond strength between the studied groups of enamel and dentin and was done using F-test (ANOVA) and Post Hoc test (Tukey). Significance level was set at the 5% level.

For observation of the Interfacial morphology, three specimens from each group were further longitudinally sectioned in a mesio-distal direction to expose the resin-tooth interface using a diamond disc at low speed handpiece under water coolant. The exposed surfaces were air-dried and smoothed with increasingly finer grit silicon carbide papers (#600, #1000, #1500, #2000) under running water. They were then etched with 37% phosphoric acid solution for 10 seconds to remove the mineral content, washed with water and immersed in 5% sodium hypochlorite for 5min to remove exposed collagen for better observation of resin tags. Finally, specimens were subjected to ultrasonic bath with distilled water for 10 min and allowed to dry overnight and then gold sputtered and observed under SEM (JEOL JSM -5300 Scanning Microscope, Japan) at x3.500 magnification (17,18).

RESULTS

In enamel, the difference in the mean values of shear bond strength ofthe four groups showed a statistically significant difference with P <0.001. Group A1 and Group A3 showed the highest mean values (35.60 and 32.60 MPa respectively). Group A2 and Group A4 showed no statistically significant difference with mean values of (25.69 and 23.78 MPa respectively). Etching enamel revealed a statistically significant increase in the mean values of shear bond strength of RelyX ultimate cement with P <0.001(Table 2).

Table (2): Comparison between the different studied groups of enamel according to shear bond strength (MPa).

	Enamel groups				F	p
	A1 (n = 10)	A2 (n = 10)	A3 (n = 10)	A4 (n = 10)		
Shear bond Strength (MPa)						
Min. – Max.	32.80 – 39.70	23.0 – 31.40	30.55 – 38.15	21.20 – 29.20		
Mean ± SD.	35.60 ± 2.51	26.69 ± 2.82	32.60 ± 2.34	23.78 ± 2.37	45.997*	<0.001*
Median	35.2	26.55	31.88	23.40		
p_1		<0.001*	0.053	<0.001*		
p_2			<0.001*	0.064		
p_3				<0.001*		

F: F test (ANOVA) *: Statistically sig. at p ≤ 0.05.

While for dentin, the difference in the mean values of shear bond strength of the four groups showed generally no statistically significant difference with P=0.053. Mean values were (21.68 MPa, 19.72 MPa, 20.73 MPa and 18.64 MPa respectively). An etching step prior to RelyX ultimate cement resulted in a non-significantly increased bond strength with P=0.302(Table 3).

Table (3): Comparison between the different studied groups of dentin according to shear bond strength (MPa).

	Dentin groups				F	p
	B1 (n = 10)	B2 (n = 10)	B3 (n = 10)	B4 (n = 10)		
Shear bond Strength (MPa)						
Min. – Max.	18.80 – 26.40	15.60 – 24.30	17.80 – 25.40	16.40 – 23.80		
Mean ± SD.	21.68 ± 2.35	19.72 ± 2.27	20.73 ± 2.73	18.64 ± 2.50	2.809	0.053
Median	21.40	19.50	20.20	17.85		
p_1		0.302	0.825	0.043*		
p_2			0.797	0.763		
p_3				0.249		

F: F test (ANOVA) *: Statistically sig. at p ≤ 0.05.

SEM micrographs of the resin-enamel interface from RelyX ultimate in total-etch mode showed thin resin tags penetrating into enamel.When applied in self-etch mode, there was absence of any penetrating resin tags and an evident hybrid layer was seen. Variolink II showed fewer and longer resin tags than RelyX ultimate in total-etch mode, while Multilink automix showed very thin, non-uniform hybrid layer with absence of any resin tags (Figure 1).

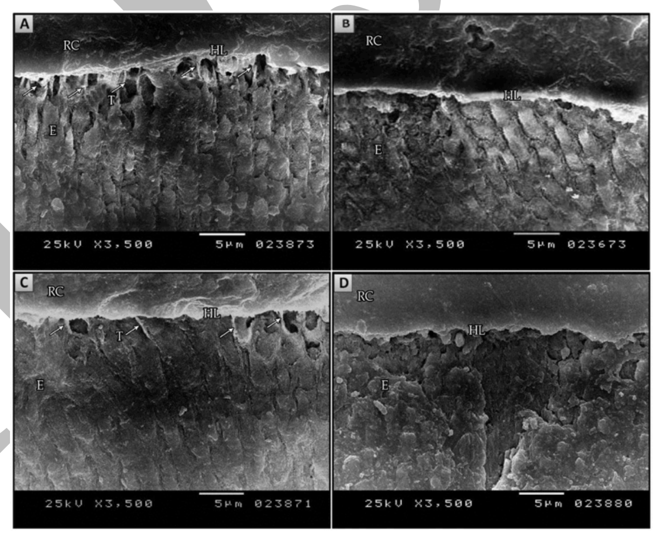

Fig.1: SEM micrographs of the resin-enamel interface formed by (A) RelyX ultimate resin cement in total-etch mode; (B) RelyX ultimate resin cement in self-etch mode; (C) Variolink II total-etch resin cement; (D) Multilink automix self-etch resin cement, x3500. RC: Resin cement, E: Enamel, HL: hybrid layer, T: Resin tags.

Resin-dentin interface showed that RelyX ultimate in total-etch mode was able to penetrate into dentin forming thicker hybrid layer with numerous, well-defined and deep resin tags. When applied in self-etch mode, very few resin tags with shallow penetration and thin uniform hybrid layer were seen. Variolink II showed variability in the length and less density of resin tags than RelyX ultimate as total-etch. Multilink automix showed thicker hybrid layer than RelyX ultimate as self-etch and few resin tags of variable shape with shallow penetration (Figure2).

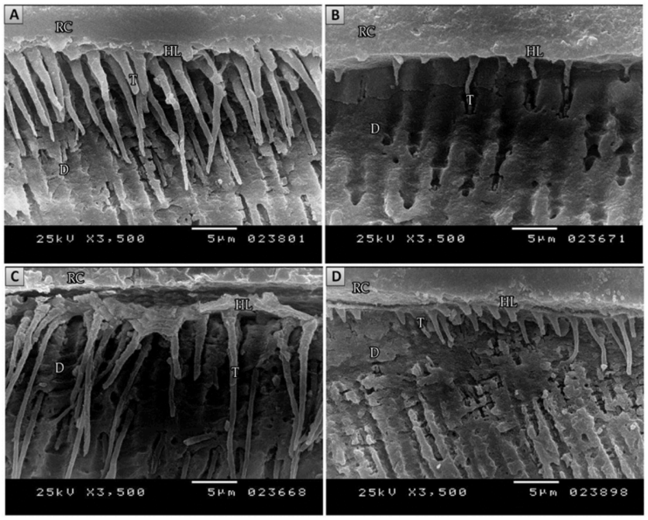

Fig.2: SEM micrographs of the resin-dentin interface formed by (A) RelyX ultimate resin cement in total-etch mode; (B) RelyX ultimate resin cement in self-etch mode; (C) Variolink II total-etch resin cement; (D) Multilink automix self-etch resin cement, x3500. RC: Resin cement, D: Dentin, HL: Hybrid layer, T: Resin tags.

DISCUSSION

The main challenge for dental adhesives is to provide an equally effective bond to two hard tissues of different nature. One of the most recent novelties in adhesive dentistry was the introduction of multi-mode adhesives that usually contain all bonding components in a single bottle and may be applied either in etch-and-rinse or self-etch bonding approaches (12).

In the present study the results of shear bond strength to enamel revealed that RelyX ultimate in total-etch mode and Variolink II total-etch resin cement had a significantly higher bond strength than Multilink automix self-etch resin cement, this comes in agreement with Hikita et al (3) and Inoue et al (5) who also reported higher enamel bond strengths when adhesive cements were combined with total-etch adhesives. These findings may be attributed to the demineralization capacity of the etching agents as studies have shown that when enamel was etched by phosphoric acid the adhesive system penetrates to a depth of 1 μm between the enamel crystallites forming micromechanical interlocking through the formation of resin tags, whereas enamel etched by components of the self-etch adhesive the adhesive only reaches a depth of 0.6-0.7 μm and is based on an inter- and intra-crystallite hybridization of enamel surface (19,20). Thus, the shallower etching pattern on enamel and subsequent reduced micro-mechanical retention might jeopardize bonding (21). The results of interfacial morphology in this study supported the bond-strength data, since more intimate micromechanical interlocking at the etched enamel surface could be achieved.

Furthermore, when enamel was etched prior to application of RelyX ultimate multi-mode adhesive cement, the results of the current study showed significant increase in the bond strength values and betterinterfacial morphology. Therefore, the null hypothesis that etching of enamel does not influence the bonding efficiency of multi-mode adhesive system was rejected. These results are in line with Erickson et al (22) and Li et al (23) who found improved bond strengths with a pre-etch step and attributed this to the degree of etching or the etch morphology achieved as etching increase the bonding area and the wettability of the adherent surface, promoting good micromechanical retention and increasing the bond strength values. However, Watanabe et al (24) stated that improvement in bond strengths after etching of enamel surfaces using phosphoric acid depends on the brand of the self-etch adhesive used, so not only the depth of enamel etching, but also the composition and mechanical properties of the self-etch adhesives might play important roles in determining bond strength

The results of this study also revealed that RelyX ultimate in self-etch mode had higher bond strength values than Multilink automix self-etch resin cement. This findings may be attributed to the composition of Scotchbond universal (multi-mode adhesive) that was used with RelyX ultimate cement, it also contains 10-MDP functional acidic monomer which has been found to interact chemically with hydroxyapatite (Adhesion-decalcification concept) (25).

MDP-containing adhesives form nanolayers at the adhesive interface, on which calcium ions released up for a highly hydrolytically stable Ca-10-MDP formation which is the most stable salt compared with other salts formed from experimental acidic monomers (26,27), so the high bond strength may be correlated to stable MDP-Ca salt deposition along with the strong hydrophobicity of the nanolayered structure that can protect the hybrid layer against hydrolytic bond degradation processes. Moreover, the polyalkenoic acid copolymer that also presents in Scotchbond universal adhesive increased the interaction of the adhesive with enamel as the carboxyl groups of the polyalkenoic acid dissociate to release protons in aqueous solutions enabling interacting in acid-base reactions. Carboxyl groups can replace phosphate ions of the substrate and form ionic bonds with Ca increasing interaction of the adhesive (28).

Bonding to dentin is more challenging due to the higher organic content, fluid pressure from dentinal tubules and the presence of a smear layer (29). In the present study the results of shear bond strength to dentin showed that RelyX ultimate in total-etch mode and Variolink II achieved better results compared to Multilink automix self-etch cement, this comes in agreement with Yin M et al (30) and Özcan M (16) who found that total-etch resin cements performed better than other self-etch and self-adhesive resin cements on dentin. These findings may be attributed to surface energy of dentin and the wettability of the resin cement on surface, asphosphoric acid removes the smear layer and increases surface roughness and thereby, the wettability of the adhesive resin (31). Interfacial morphology in this study showed deep resin tags penetrating the etched dentin surfaces as phosphoric acid opened dentinal tubules, peritubular dentin is removed, which leads to enlargement of the dentinal tubules, resulting in deeper penetration of the resin monomers into the exposed collagen network and mechanical interlocking with dentin with improved bonding penetration and more effective surface conditioning (32).

The relationship between resin tags and the bond strength remains a controversial issue, in the current study, numerous tags that were seen when RelyX ultimate was used in total-etch mode may be responsible for the increased bond strength values than Variolink II with fewer and longer resin tags, suggesting that the length of the resin tags did not contribute much to bond strength, this comes in agreement with Yoshiyama et al (33) who stated that the length of resin tags seems not to play an important role regarding the bond strength. According to Gwinnett (32), resin tags can contribute approximately one-third of the total shear bond strength. However, Tao and Pashley (34) reported that no link existed between the bond strength and the formation of the resin tags.

The results of the current study showed that RelyX ultimate cement had a non-significant increase in shear bond strength values when applied to dentin in the total-etch mode. Therefore, the null hypothesis that etching of dentin does not influence the bonding efficiency of multi-mode adhesive system was accepted, also the interfacial morphology showed uniform hybrid layer, numerous, well-defined and deeper resin tags compared to respective self-etch counterpart, this comes in agreement with Wagner et al (13) who found improved bond strength and penetration pattern when Scotchbond universal adhesive was applied as a total-etch on dentin. However, this finding contradicts with previous studies (9, 35) which revealed that etching of dentin prior to the application of self-etch adhesivesdecreased the bond strength values. They correlated this to collapse of collagen during the drying process after rinsing the etchant that lead to incomplete infiltration of the deeply demineralized collagen network by the bonding resin which has been observed as voids or micro porosity beneath and within the hybrid layer (36). This shortcoming has been overcome in Scotchbond multi-mode adhesive through the addition of low viscosity monomers like HEMA, that increase the affinity to the hydrophilic wet collagen network (13).

The results of the interfacial morphology in this study showed that Multilink automix demonstrated a better ability to remove the smear layer and create thicker hybrid layer on dentin than RelyX ultimate in self-etch mode. This finding may be attributed to the etching aggressiveness of self-etch adhesive systems as Multilink primer has PH of 1.9 (intermediately strong) (37), whereas Scotchbond universal adhesive has PH of 2.7 and considered as ultra-mild adhesive (14).

However, Multilink automix along with the intermediately strong adhesive used in this study showed lower bond strength values than RelyX ultimate in self-etch mode, this comes in agreement with other studies (38, 39) which revealed that higher aggressiveness of self-etch adhesives is usually not related to higher bond strengths. This finding could be correlated to presence of water in the dentinal tubules, some of the acidic monomers of the stronger self-etch adhesives retain their acidity and continue etching and result in incomplete polymerization of the subsequently applied resin (33). On the other hand, Yoshihara et al (40) found that the MDP monomer presents in the Scotchbond universal adhesive not only has the ability to chemically bond to hydroxyapatite, but also to self-assemble into nanolayers that protect the formed hybrid layer from hydrolytic degradation.

CONCLUSIONS

From this study it was concluded that:

1) An etching step prior to multi-mode adhesive resin cement significantly increased the enamel bond strength values, while for dentin there was a non-significantly increase in bond strength with improved penetration pattern.
2) The length of resin tags formed by total-etch adhesives seems not to play an important role in determining its bond strength.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest

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