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Effect of Dentin Surface Modification on the Microtensile Bond Strength of Self-Adhesive Resin Cements

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Abstract

Purpose: To explore the potential to modify human dentin surface as a means of improving the microtensile bond strength (μTBS) of resin cement to dentin.
Materials and Methods: Sound human molars were collected, and their occlusal surfaces were ground flat to expose polished dentin. Indirect composite resin cylinders were cemented to the teeth with RelyX Unicem or G-Cem self-adhesive cements following dentin surface treatments: 6.5% grape-seed extract, 5% glutaraldehyde, or 25% polyacrylic acid and control (no pretreatment). After 24 hours, the teeth were sectioned into beams to produce a cross-sectional area of 1.0 mm^2. Specimens of each group (n = 25) were individually mounted on a jig and placed on a tensile testing machine. A tensile force was applied to failure at a 1 mm/min crosshead speed.
Results: The use of polyacrylic acid on dentin prior to cementation with RelyX Unicem resulted in a statistically significant increase in μTBS compared to the control group ($p = 0.0282$). Polyacrylic acid ($p = 0.0016$) or glutaraldehyde ($p = 0.0043$) resulted in a statistically significant increase in μTBS of G-Cem to dentin when compared to the control group. Treatment with grape-seed extract did not result in a statistically significant increase in μTBS for either cement ($p > 0.05$).
Conclusions: Priming dentin surfaces prior to the use of self-adhesive resin cements may be a promising means of improving μTBS. In addition, it was concluded that the results of this study are material dependent as well as being dependent of the type of dentin primer.

Self-adhesive cements have been used in a large range of prosthetic dentistry applications. Ease of application, decreased postoperative sensitivity, and lower susceptibility to moisture[1,2] are the main benefits of this material.

In contrast to conventional resin cements, where the adhesion to the dentin structure is achieved by surface pretreatment with etching followed by application of a bonding system to form a hybrid layer (HL) between luting resins and dentin, self-adhesive cements partially remove the smear layer through acidic monomers producing a micro-mechanical retention to the tooth structure.[3] The reaction between phosphoric acid monomers and hydroxyapatite of the dental hard tissues may also be due to chemical retention.[3,4]

Different types of commercially available materials contain diverse acid-functionalized monomers.[5] Therefore, the ability of the cement to adhere to dental substrate may be component dependent.[6] Poor adhesion may occur due to the limited etching potential of the self-etching system, which could impair the proper infiltration of the cement into dentin.[4,7-9] As a consequence, these systems present lower bond strengths than conventional multistep resin cements do.[4,7-11]

Studies have proposed different pretreatments to enhance the bond strength of self-adhesive cements.[6,12-14] Some are not effective due to smear layer removal. Self-adhesive cements are not able to infiltrate completely into the irregularities created by the agents, producing a detrimental effect on the bond strength of the materials to dentin.[6] However, polyacrylic acid (PAA), a mild acidic agent, was effective in improving the bond strength of self-adhesive cement.[12] The acid partially removes the smear layer,[12] leaving free calcium and phosphate ions on the dentin surface, providing a better chemical reaction with self-adhesive cements.[6,12,13]

It is hypothesized that the quality and longevity of the dentin bond may be improved by improving collagen properties. The biomodification of tooth dentin is a novel approach proposed to improve the biomechanical and biochemical properties of the tissue for restorative purposes. A variety of cross-linking agents, such as glutaraldehyde (GD) and proanthocyanidin-rich extracts, have been shown to effectively increase the mechanical properties of the dentin organic matrix[15-17] and superficial undemineralized dentin. Hence, specific cross-linking agents could improve the cohesive forces of the smear layer, cross linking the collagen in the smear layer and/or underlying dentin.[17,18]

The purpose of this study was to evaluate the effectiveness of surface primers in improving the microtensile bond strength (μTBS) of an indirect composite resin to sound dentin using self-adhesive resin cements. Dentin surface primers assessed included PAA, which partially removes the smear layer, as well as GD and grape-seed extract (GSE), both of which chemically modify the collagen. The null hypothesis was that the application of different dentin primers would not affect the μTBS of self-adhesive cements to dentin.

Materials and methods

Extracted sound human molars collected from dental offices were kept in a 0.1% thymol solution, cleaned, and kept frozen until use. The use of human teeth in this study was considered exempt by the Institutional Review Board at the University of Illinois at Chicago (protocol #2006–0229). The occlusal surfaces were ground flat, perpendicular to the long axis of the teeth, with #180, 320, and 600 grit silicon carbide paper (Buehler, Lake Bluff, IL) under running water to expose flat middle dentin.

Teeth were randomly assigned into two groups (n = 20) according to the cement used, RelyX Unicem (RXU) (3M ESPE, St. Paul, MN) or G-Cem (GC America, Alsip, IL). Each group was further subdivided into four groups (n = 5) according to the dentin primer:

1- Control group (no treatment): teeth were rinsed with distilled water for 5 seconds and blotted dry with an absorbent paper (KimWipe, Kimberly-Clark Corporation, Irving, TX);
2- GSE group: 6.5% GSE (MegaNatural gold grape-seed extract, Polyphenolics, Madera, CA) solution (pH ~7.2) was applied with an applicator for 2 minutes. The GSE was rinsed off with distilled water for 10 seconds, and the teeth were blotted dry with an absorbent paper;
3- GD group: 5% GD solution (pH ~ 7.2) (Fisher Biotech, Fair Lawn, NJ) was applied to the teeth for 2 minutes and then rinsed off with distilled water for 10 seconds. The teeth were blotted dry with an absorbent paper (KimWipe);
4- PAA group: 25% PAA (Ketac Conditioner, 3M ESPE, Seefeld, Germany) was applied with an applicator for 10 seconds. The PAA was then rinsed off with distilled water for 10 seconds, and the teeth were blotted dry with an absorbent paper.

In all groups care was taken to avoid desiccation of the dentin.

Laboratory composite resin cylinders (11-mm diameter, 4-mm thick) (Tescera ATL, Bisco, Schaumburg, IL, USA) were manufactured in a split aluminum mold. The blocks were processed according the manufacturers' recommendations in a specific light-curing unit under pressure and heat (TESCERA ATL™, Bisco Co. Schaumburg, IL, USA). The surface of each composite resin cylinder was sandblasted with 50-μm aluminum oxide particles for 5 seconds, rinsed with distilled water, and air dried.

These cylinders were cemented to the teeth following each cement manufacturers' directions. A 100-g occlusal load was applied to the crown. The specimens were light polymerized with a curing unit (Optilux 501, SDS Kerr, Middleton, WI) for 10 seconds after which time a hand instrument was used to scale off the excess cement. The specimens were then light polymerized for an additional 20 seconds from each direction: occlusal, buccal, lingual, mesial, and distal. The specimens were then stored in distilled water at 37°C for 24 hours.

After storage time elapsed, the specimens were sectioned perpendicular to the adhesive-tooth interface into beams (~1.0 × 1.0 mm) using a slow-speed diamond wafering blade (Buehler-Series 15LC Diamond, Buehler) under constant water coolant. Electronic digital calipers (Thermo Fisher Scientific Inc., Pittsburgh, PA) were used to measure the beams so that the cross-sectional area of the interface could be calculated. Five beams were selected from each tooth, totaling 25 beams for each test condition. The use of beams as experimental units can be supported by previous studies that evaluated μTBS of resin cement to teeth.[2,11-13] To perform the microtensile bond test, each beam was affixed with cyanoacrylate adhesive (Loctite Superglue Gel, Henkel Consumer Adhesives, Avon, OH) and tested to failure using a microtensile tester (Bisco) at a 1 mm/min crosshead speed. The peak load was recorded, and μTBS values were obtained by dividing the peak break force by the cross-sectional area (mm^2) of the beam. Data were expressed in MPa.

The μTBS data were statistically analyzed by two-way ANOVA ($\alpha = 0.05$). Type of cement (two levels) and dentin pretreatment (four levels) were evaluated to determine if these factors influenced μTBS. Post hoc Fisher's test was used to analyze each type of self-adhesive cement and to determine whether various combinations of cement and primers resulted in statistically significant differences.

Table 1 Results of the microtensile bond strength [mean (standard deviation)] evaluation of self-adhesive resin cements bonded to dentin following different dentin treatment

	Microtensile bond strength (MPa)			
	Dentin surface treatment			
Resin cement	No pretreatment	PAA	GSE	GD
Rely-X Unicem	12.2 (5.1)a	16.6 (9.3)b	13.0 (7.6)a	13.9 (5.0)a
G-Cem	11.4 (6.5)a	19.5 (10.0)b	8.3 (4.3)a	18.7 (10.1)b

Different letters indicate statistically significant differences in each row ($p < 0.05$).

Results

Two-way ANOVA indicated a statistically significant interaction between the factors (dentin treatment vs. cements, $p = 0.0282$). The dentin primers significantly affected the μTBS ($p < 0.0001$), while no differences were observed between the resin cements ($p = 0.6323$; Table 1). When G-Cem was used, PAA and GD treatments resulted in μTBS greater than control ($p = 0.0016$ and $p = 0.0043$, respectively). PAA pretreatment also significantly increased the μTBS of RXU to dentin ($p = 0.0282$), while GD had no statistically significant effect on the bond strength ($p = 0.3749$). GSE did not affect the μTBS for both resin cements (RXU, $p = 0.6665$; G-Cem, $p = 0.2542$). Some specimens debonded prematurely during preparation; however, pretest failures were not included in the statistical analysis.[11]

Discussion

The findings of this study indicate that the use of a specific dentin primer modified the μTBS of self-adhesive cements. It appears that only certain combinations of dentin pretreatment primers and luting materials result in improved μTBS. PAA treatment resulted in statistically significantly higher μTBS for both cements, while GD only significantly increased the μTBS for G-Cem. Pretreatment with GSE did not result in a statistically significant change in μTBS regardless of whether RXU or G-Cem was used. Therefore, the null hypothesis was partially rejected.

There are a variety of possible reasons for these findings. The primers' effect on the smear layer may have influenced the results. Manufacturers of self-adhesive cements claim that they are capable of removing the smear layer. Studies have shown different results.[4,7-9] Though RXU and G-Cem both have acidic pHs (2.1 and 2.7, respectively), neither has been shown capable of smear layer dissolution, dentin demineralization, and HL formation.[5,9] In this study both cements showed similar bond strength values when compared to no dentin priming. These results are similar to findings of different studies that evaluate the bond of self-adhesive cements to untreated dentin.[2,7,11,12] However, the bond strength of these cements are lower than conventional multi-step cements.[4,7,11]

The PAA treatment resulted in a statistically significantly higher μTBS for RelyX Unicem and G-Cem. Both cements contain glass ionomer filler particles in their composition, suggesting that some characteristics of glass ionomer cement can emerge inevitably to a certain extent.[6,11,19] The mild acid agent partially removed the smear layer, leaving the dentin mineral phase, thus enhancing the chemical reaction between the cement and the substrate.[6,12,13,20,21] The functional carboxyl ion groups present in PAA formulation can form a multiplicity of hydrogen bonds.[8,22] Additionally, the partial cleaning of dentinal tubules may increase water presence at the interface from moisture present at the pulpal tissue. Great effort was taken to keep the specimens hydrated during preparation and restoration. Moisture on the dentin surface can play a role in the adhesion of self-adhesive cements.[1,2,6] A previous study observed an increase in the bond strength of RXU to dentin when pulpal pressure was simulated, while it did not affect the dentin bond of G-Cem.[8] RXU bond strength may improve due to increasing the hydration state of dentin, because phosphoric acid esters may require water to become ionized and interact with dentin.[1] In this study, both cements showed enhanced bond strength to the substrate, suggesting that the increased water in the environment may have optimized the acid/base reaction and improved bond strength.

Pretreatment with GSE did not result in a statistically significant change in μTBS for both cements, while GD showed an increased μTBS for G-Cem cement. Because the pH of the GD and GSE was adjusted to approximately 7.2 in this study, it is unlikely that either appreciably reduced the thickness of the smear layer. A possibility is that the behavior of the GD and GSE molecules may have played a role. GD and GSE are capable of increasing the number of collagen cross-links in dentin, resulting in improved mechanical properties of the tooth.[15,16] GD increases the number of collagen cross-links via the formation of a Schiff's base, which occurs when an aldehyde of GD reacts with a collagen amino group of lysine or hydroxylysine. GD may also exert its effect when aldol condensation occurs between two adjacent aldehydes.[23] In contrast, proanthocyanidin, a polyphenol obtained from GSE, is believed to interact with collagen proteins by covalent, ionic, hydrogen bonding, and hydrophobic interactions.[15,24,25] During the pretreatment with GD or with GSE, the agent must make contact with collagen within the dentin. While no changes to the mechanical properties of GSE- and GD-treated mineralized dentin have been shown at the micro-scale,[15] recent nano-scale findings showed increased mechanical properties at the dentin surface exposed to both agents (unpublished data). It had been shown that GD diffuses through the mineralized dentin,[23,26] but GSE's effect may be restricted to the very superficial layer, due to the high molecular weight of proanthocyanidin molecules.[24] This would explain the finding in this study that GSE primer did not improve the μTBS to a statistically significant degree, regardless of the cement used. While pretreatment with GD resulted in a statistically significant improvement when used in combination with G-Cem, this was not the case with RXU. It is possible that this was due to interaction between the primer agent and the cement. This is an area that deserves further study. Such interaction may, for example, result in improved surface contact or optimization of chemical reactions.

Strengths of this study include the fact that every effort was made to standardize the experimental process. A thoughtfully planned pilot study was carried out to develop an effective experimental design. While in vitro studies are an excellent starting point for exploring new treatments and techniques, they are not necessarily representative of what happens intraorally. Because attempting to modify tooth structure, rather than simply improving upon or developing new restorative materials, is a new area of interest in restorative dentistry, further research is necessary. Such agents like PAA and GD may be used as a dentin primer to improve the bond strength of self-adhesive cements. Additional studies are necessary to understand any long-term benefits. In addition, altering variables such as cross-linker application time, cross-linker pH, or different combinations of pretreatment and cement are all worth evaluating further.

Conclusions

Within the limitations of this in vitro study the following conclusions were made:

1. The use of PAA increased the μTBS of G-Cem and RelyX to dentin. GD increased the μTBS of G-Cem to dentin, and

GSE did not show a statistically significant effect on the μTBS.
2. Priming the dentin prior to cementation of indirect resin restorations with self-adhesive cements may be a promising means of improving the μTBS.

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