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Bond Strength of Adhesive Luting Agents to Caries-affected Dentin

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Clinical Relevance

Caries-affected dentin could remain during conservative preparation for indirect restorations. However, the reduced bond strength of adhesive luting agents to this altered substrate might affect the longevity of restorations.

SUMMARY

Introduction: The aim of this study was to evaluate the bond strength of adhesive luting agents to caries-affected dentin (CAD). Methods: Forty human molars were sectioned to create dental slices presenting exposed occlusal dentin. Half of the samples were submitted to eight caries-induction demineralizing/mineralizing cycles. The pH-cycling model consisted of three hours in a demineralizing solution followed by 45 hours of immersion in a mineralizing solution. Dentin hardness was measured before and after the pH cycling. Resin cement cylinders were built up over the dentin surface using RelyX Unicem or RelyX ARC/Scotchbond Multipurpose Plus. The cement cylinders were submitted to shear load, and the data were analyzed using two-way analysis of variance (ANOVA) and Tukey test ($p<0.05$). Hardness data were also submitted to two-way ANOVA and Tukey test ($p<0.05$). The relationship of hardness vs bond strength was assessed via nonlinear regression analysis. Results: Sound dentin (tested and used in caries induction) showed similar values of hardness and were superior to CAD. Both resin cements showed higher bond strength to sound dentin than to CAD. Independent of substrate, RelyX ARC showed the highest values of bond strength. A positive linear relationship between dentin hardness and bond strength was observed for both cements evaluated. Conclusions: The adhesive luting agents evaluated showed lower bond strength to CAD.

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Introduction

Improvement of adhesive luting systems and ceramic materials has resulted in more conservative preparations for indirect restorations, while the cavity preparation can be limited only to caries removal and eliminating retentive areas. In this approach, the restoration is retained mainly by bonding procedures. The goal of caries excavation is to eliminate the caries-infected dentin, which presents as an irreversibly demineralized and denatured substrate.[1,2] To preserve the dental tissue, the underlying partially preserved and remineralizable dentin (caries-affected dentin

[CAD]) is maintained.[3-5] Significant alterations of mineral content have been described for CAD, where the intertubular dentin presents lower mineral content than normal dentin, and where acid-resistant minerals are observed obliterating the dentin tubules.[6-8] The obliteration of tubules can interfere with resin infiltration, thus preventing tags during bonding procedures.[9] Conversely, the lower mineral content of intertubular dentin in CAD permits deeper etching of this substrate.[10]

Considering the maintenance of CAD during conservative cavity preparations that ensure the receiving of indirect restorations, proper bonding between the restorative material and CAD is fundamental to the success of the procedure. For many years, resin cements were used with adhesive systems to lute metal-free indirect restorations. However, multiple-step protocols of cementation increase the technical sensitivity and clinical time required for the procedure.[11] Furthermore, incompatibility between simplified adhesives and resin cements has been an issue when the chemical activation of cement polymerization is predominant.[12] Thus, simplified luting agents are gaining in popularity in such a context.

Self-adhesive resin cements (SARCs) have been developed to simplify clinical procedures and to overcome the technique sensitivity of multiple-step systems. According to manufacturers, SARCs require no pretreatment of the dental surfaces and are applied in a single clinical step.[13,14] The bonding mechanism of SARCS is attributed to a chemical reaction between phosphate methacrylates and hydroxyapatite as well as to the infiltration of these materials into the tooth tissues.[15,16] Reduced bond strength of adhesive systems to CAD has been demonstrated,[6,8,17] but little information is available about the bonding of SARCs to this clinically relevant substrate. One of the difficulties in evaluating adhesive systems' bonding to CAD involves obtaining a standardized substrate for the bond-strength studies. Thus, the artificial induction of caries has been used to permit the standardization of the bonding substrate.

The aim of this study was to evaluate the bond strengths of a conventional resin cement and a SARC to artificially created CAD. The null hypotheses tested were as follows: 1) no difference exists between the shear bond strengths of the evaluated luting agents to sound and artificially created CAD; 2) no significant correlation exists between the hardness of the substrate and bond strength; and 3) the bond strength of the evaluated resin cements to both substrates is similar.

METHODS AND MATERIALS

Noncarious human third molars stored in 0.05% thymol saline solution for no more than six months were used in this study. The occlusal surfaces were ground flat using 180-, 320-, and 600-grit silicon carbide paper under running water to remove the enamel and expose a flat dentin surface. A section was performed parallel to the occlusal surface and 2 mm below the cement-enamel junction using a water-cooled slow-speed diamond saw (#7020, KG Sorensen, Barueri, Brazil), while the roots were discarded. An adhesive tape (4×5 mm) was bonded over the occlusal dentin followed by the application of two coats of an acid-resistant, fast-drying nail varnish (Colorama Maybelline, São Paulo, Brazil) on all specimen surfaces. After the tape removal, a window of 20 mm^2 was obtained on the occlusal dentin surface.

Ten Vickers hardness indentations were made on the ground surface under a load of 50 g for a 10-second dwell time (HMV-2, Shimadzu, Tokyo, Japan). The Vickers hardness number (VHN, kgf/mm^2) for each specimen was recorded as the average of the 10 readings. Specimens that presented VHNs that differed by more than 5% from the mean of all specimens were discarded. Forty specimens were selected, and half were submitted to caries-induction demineralization/mineralization cycles. The demineralization/mineralization cycle was characterized by a three-hour immersion of specimens in a demineralizing solution (156.25 mL/tooth) followed by a 45-hour immersion in a mineralizing solution (78.125 mL/tooth). Specimens were submitted to eight cycles of 48 hours; the demineralizing solution was renewed after the fourth cycle, and the mineralizing solution was renewed before the beginning of each new cycle.[17] The compositions of the solutions are listed in Table 1. After demineralization/ mineralization cycling, new Vickers hardness readings were performed on the dentin surfaces of the cycled specimens. Hardness data were analyzed by two-way analysis of variance (ANOVA) and Tukey test ($p<0.05$).

A conventional RelyX ARC (3M ESPE, St Paul, MN, USA) and a self-adhesive RelyX Unicem (3M ESPE) were used in this study. For the RelyX ARC, the adhesive system (Scotchbond Multipurpose Plus, 3M ESPE) was applied over the dentin surface (sound and CAD) before insertion of cement. Thus, the substrate was etched with 35% phosphoric acid

Table 1: *Composition of Solutions Used to Induce Caries*

Solution	Composition
Demineralizing (pH = 4.5)	2.2 mM calcium ($CaCl_2$)
	2.2 mM phosphate (NaH_2PO_4)
	0.05M sodium acetate
	0.05M acetic acid
	1 ppm fluoride (NaF)
Mineralizing (pH = 7.0)	1.5 mM calcium ($CaCl_2$)
	0.9 mM phosphate (NaH_2PO_4)
	0.15M KCl
	0.1M Tris buffer
	10 ppm fluoride (NaF)

(Scotchbond etchant, 3M ESPE) for 15 seconds followed by rinsing of acid. Excess water was removed with absorbent paper, and the adhesive system primer was applied over the wet dentin. After five seconds, the primer solvent was volatilized with a gentle air stream. The adhesive was applied and light-cured for 25 seconds. No previous treatment of the substrate was performed for RelyX Unicem.

Polyvinyl siloxane molds (Aquasil Extra Low Viscosity, Dentsply DeTrey, Konstanz, Germany) containing four cylindrical cavities (1 mm in diameter, 2 mm high) were placed on the dentin surface. The mold cavities were filled with one of the cements, which were light-cured for 20 seconds. All light curings were performed using a light-emitting diode unit (Radii Cal, SDI, Bayswater, Victoria, Australia) with an irradiance of 800 mW/cm^2.

After 24 hours of storage (37°C at 100% humidity), shear bond tests were conducted using a universal testing machine (Instron 5565, Instron, Canton, MA, USA). A thin steel wire (0.2 mm in diameter) was looped around each cylinder, and a shear load was applied to the base of the cylinder at a crosshead speed of 0.5 mm/minute until failure. The average load at failure of the four cylinders was recorded as the microshear bond strength (MPa) for that specimen. Bond strength data were analyzed using two-way ANOVA and Tukey test. The factors evaluated were "cement" and "dentin." The relationship between hardness and bond strength was assessed by nonlinear regression analyses, with bond strength as a dependent variable. All analyses were conducted at a significance level of $p<0.05$.

RESULTS

For the hardness data, ANOVA did not show a significant effect for the factor "cement" ($p=0.232$) and for the interaction between the factors ($p=0.429$). These results showed that both cements evaluated were used in similar dentin. Only the factor "dentin" showed a significant effect ($p<0.001$). The dentin used for caries induction and those tested as sound dentin showed similar hardness values that were higher than those observed for CAD. The results are displayed in Table 2.

Table 2: *Means (SD) of Hardness in VHN*[a]

Resin Cement	Dentin		
	Sound	Moment of CAD Induction	
		Before	After
RelyX ARC	49.0 (1.2)	51.7 (3.3)	15.5 (3.5)
RelyX Unicem	49.6 (3.0)	49.5 (3.4)	13.7 (3.1)
Pooled average	49.3 (2.2)^A	50.6 (3.4)^A	14.6 (3.3)^B

[a] *For pooled average, distinct letters indicate statistical difference (p<0.05).*

For the bond strength data, ANOVA showed a significant effect for the factors "cement" ($p=0.002$) and "dentin" ($p<0.001$) but not for the interaction between the factors ($p=0.209$). The cement RelyX ARC showed higher bond strength than RelyX Unicem did for both substrates. Independent of cement, the highest bond strength values were observed for sound dentin. The results are displayed in Table 3. The relationship between hardness and bond strength followed a significant positive linear behavior for both RelyX ARC ($R^2 = 0.80$; $p<0.001$) and RelyX Unicem ($R^2 = 0.89$; $p<0.001$). A predominance of adhesive failures was evident.

DISCUSSION

Despite the importance of CAD in restorative procedures, most studies that evaluate the bond strength of adhesive materials to dental substrates are performed using sound dentin. The standardization of CAD *in vitro* studies is difficult to determine, and differences in the caries removal and/or caries detection methods can compromise the comparison of results obtained in different studies.[18-20] Even when using proper methods for detecting and removing caries, a heterogeneous substrate can remain after removal of the caries-infected dentin.[21] In some cases, the presence of small sites of CAD surrounded by sound dentin has been observed. Because the sizes and shapes of these sites are often irregular,[22] standardizing the sample preparation for bonding tests is too difficult.

This study artificially induced CAD through demineralizing/mineralizing cycling. This method has been previously described and results in a substrate with similar mineral content to that

Table 3: *Means (SD) for Bond Strength in MPa*[a]

Resin Cement	Dentin		Pooled Average
	Sound	CAD	
RelyX ARC	23.6 (6.8)	8.5 (2.9)	16.0 (9.3) A
RelyX Unicem	16.0 (3.7)	5.0 (1.8)	10.5 (6.4) B
Pooled average	19.8 (6.6) a	6.7 (2.9) b	

[a] *For pooled average, distinct letters (uppercase for cement, lowercase for dentin) indicate statistical difference ($p<0.05$).*

observed in natural CAD.[2,8,17] Natural CAD presents as a hypomineralized tissue because of the cycles of demineralization and remineralization over a long period.[8] This process results in dissolution of mineral content, which precipitates into dentinal tubules, thus creating crystal logs.[7,23] In contrast, it has been demonstrated that short periods of artificial caries induction do not permit the formation of crystallites in the dentinal tubules.[8] Despite this difference, similar values of bond strength between natural and artificial CAD have been reported, though a lower variability of bond strength was observed for artificially induced CAD.[8]

In the present study, the hardness of the evaluated dentin substrate was measured before the induction of CAD. The sound dentin and the dentin used for caries induction showed similar dentin hardness values, demonstrating homogeneity in mineral content between the samples used. Meanwhile, the samples submitted to demineralization/remineralization cycling presented the lowest values of bond strength. These results were expected based on the longer time the samples spent in the demineralization solution. Reduction of dentin hardness also resulted in lower bond-strength values for both adhesive luting agents, and a positive significant correlation was observed between these two measurements. Thus, the first and second hypotheses of the study were rejected.

RelyX ARC was used in the present study, associated with the three-step etch-and-rinse adhesive Scotchbond Multipurpose Plus. Previous studies have also demonstrated reduced bond strength of etch-and-rinse adhesives to CAD when compared with their bond strength to sound dentin.[6,8,17] The presence of crystals in the dentin tubules has explained this reduction, as they reduce the extension of the resin tags.[6] Despite the fact that the artificially induced CAD does not present these crystals, the presence of tags in the dentinal tubules is weakly related to bond strength. Thus, the deeper demineralization that phosphoric acid promotes on more porous CAD can explain the results.[10] A higher discrepancy is expected between the depth of dentin demineralization and adhesive resin infiltration for CAD, resulting in a thicker layer of an unprotected mineral-depleted collagen at the base of the hybrid layer.[24,25] This mineral-depleted layer can act as weak link during shear testing and therefore reduce the bond strength.[25]

Previous studies showed the absence of resin tags and evident dentin demineralization for the self-adhesive resin cement RelyX Unicem when applied on sound dentin.[26,27] The high viscosity and neutralization effect during the setting of this material have been related to this limited capacity to effectively diffuse and decalcify the dentin.[26] Thus, the main bonding mechanism of RelyX Unicem to dentin is probably due to a chemical reaction between the phosphate methacrylates and hydroxyapatite.[15] Considering the importance of the presence of calcium for the proper boding of Unicem to dentin, the lower mineral content of CAD can explain the results. Independent of the substrate evaluated, ARC showed higher bond strength did than Unicem, rejecting the study's third hypothesis. The formation of an effective hybrid layer for ARC, different from that of Unicem, can explain these results.

This study showed that the reduction in hardness that artificial caries induction promotes resulted in the lowest bond strength values for a conventional resin cement and a SARC. The pH-cycling method used in this study simulates *in vivo* physicochemical variations involved in the caries progression. However, the model of artificial caries induction used in this study presents some limitations. In addition to the absence of crystals in dentinal tubules when this method of caries induction is used, the proteinase releases of the microorganisms have an important role in collagen degradation.[28,29] Considering the importance of these collagen alterations and the standardization of the bonding substrate, the use of a microbiologic model for caries induction can produce an ideal model for evaluating bonding to CAD. Thus, further studies for evaluating the bond strength of a SARC to CAD must be conducted to confirm the present study's outcomes.

CONCLUSIONS

Within the current study's limitations, the following conclusions can be made:

- The artificial CAD showed lower hardness than did sound dentin.
- Both evaluated adhesive luting agents showed lower bond strength to CAD than did sound dentin.
- Independent of the substrate, RelyX ARC showed higher values of bond strength than did RelyX Unicem.

Conflict of Interest

The authors of this manuscript certify that they have no proprietary, financial, or other personal interest of any nature or kind in any product, service, and/or company that is presented in this article.

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