Effect of Smear Layer Thickness and pH of Self-adhesive Resin Cements on the Shear Bond Strength to Dentin

Abstract

Context: There are concerns in relation to the bonding efficacy of self-adhesive resin cements to dentin covered with the smear layer. Aims: This study aims to evaluate the effect of smear layer thickness and different pH values of self-adhesive resin cements on the shear bond strength to dentin. Materials and Methods: The dentin on the buccal and lingual surfaces of 48 sound human premolars were abraded with 60- and 600-grit silicon carbide papers to achieve thick and thin smear layers, respectively. The samples were divided into three groups (n = 16) based on the cement pH: Rely-X Unicem (RXU) (pH < 2); Clearfil SA Luting (CSL) (pH = 3); and Speed CEM (SPC) (pH = 4.5). In each group, composite resin blocks were bonded to the buccal and lingual surfaces. After 24 h, the shear bond strength values were measured in MPa, and the failure modes were evaluated under a stereomicroscope. Statistical Analysis: Data were analyzed with two-way ANOVA and post hoc least significant difference tests (P < 0.05). Results: Cement pH had a significant effect on the shear bond strength (P = 0.02); however, the smear layer thickness had no significant effect on the shear bond strength (P > 0.05). The cumulative effect of these variables was not significant, either (P = 0.11). Conclusion: The shear bond strengths of SPC and CSL self-adhesive resin cements were similar and significantly lower than that of RXU. The smear layer thickness was not a determining factor for the shear bond strength value of self-adhesive resin cements.

Keywords: pH, resin cement, self-adhesive, shear bond strength, smear layer

Introduction

Resin cements are the materials of choice for cementation of indirect esthetic restorations. In general, the resin cements are applied after acid etching and use of an etch-and-rinse adhesive system, a process which significantly increases the bond strength to tooth structures. However, this technique is highly technique-sensitive and complex because it consists of several steps and the bonding efficacy might be compromised due to different factors, including incomplete penetration of the adhesive into the demineralized dentin, inadequate exposure of collagen, poor seal of dentinal tubules, inadequate polymerization of the adhesive, leakage of some adhesive systems, and decreased diffusion of resin monomers, which might result in postoperative sensitivity and failure of the indirect restoration.

Nowadays, due to the difficulty of the use of etch-and-rinse systems, there is a tendency to make the clinical steps easier, which has resulted in the introduction of self-adhesive resin cements. In this new generation of resin cements, the adhesive and the cement have been incorporated into one step, simultaneously resulting in decalcification and penetration into the matrix and negating the need for prior preparation of the root surface.

Piwowarczyk et al. have reported that the bond strength of this luting system to dentin was comparable to that of the commonly used luting systems. In addition, Bitter et al. showed higher bond strength of a self-adhesive system to intracanal dentin compared to conventional resin systems. However, there is still controversy over the bonding efficacy of these simplified cements to dentin covered with the smear layer.

Previous studies have shown that complete removal of the smear layer by acid etching results in an increase in the bond strength of self-adhesive cements to enamel; however, contrary to expectations, acid etching decreases the bond strength to dentin. In addition, it has been shown that use of...
weaker acid solutions such as ethylenediaminetetraacetic acid and polyacrylic acid, which remove the smear layer partially and preserve some of the mineral phase of dentin, results in an increase in the odds of chemical reaction between the self-adhesive cement and the matrix.[12-14] Recent studies have shown that self-adhesive cements can take part in reactions with the superficial dentin, resulting in partial demineralization of the smear layer and formation of short resin tags.[15-17] Under clinical conditions, surface roughness and the thickness of the smear layer will be different depending on the type of the rotary instrument used.[18,19] A thick smear layer might affect the ability of self-adhesive systems to penetrate into the intact mineralized dentin; in addition, premature neutralization of the adhesive by the buffering components of the smear layer might prevent decalcification of the dentin surface, which is a prerequisite for the exposure of collagen fibers.[20] Previous studies have shown that the penetration depth of self-etch adhesives into the subsurface dentin depends on the acidity or pH of the self-etch system.[21,22] However, the chemical composition of self-adhesive resin cements and their reaction with dental tissues differ from that of self-etch adhesive systems.[23] Therefore, the aim of the present study was to evaluate the effect of smear layer thickness and different pH values of self-adhesive resin cements on the shear bond strength to dentin.

Materials and Methods

Forty-eight sound human premolars with mature apices, extracted from patients’ 15-25 years of age for orthodontic reasons, were selected. The teeth were cleaned of all the calculus and debris by an ultrasonic device and stored in 0.5% chloramine-T trihydrate solution (Merck, Hamburg, Germany) for up to 3 months until mounted in self-cured acrylic resin for the purpose of the study.

A diamond saw (Ortho Technology, the Hogne, Netherlands) in a high-speed handpiece (NSK, Tokyo, Japan) was used to expose dentin by removing enamel from the facial and lingual surfaces. Lingual surfaces of teeth were polishing with 600-grit silicon carbide (SiC) under running water for 1 min to produce flat bonding surfaces with thin smear layers. The buccal surfaces were polished similarly with 60-grit SiC paper to produce surfaces with thick smear layers.[24,25] It should be noted that to ensure the similarity of the smear layer thickness on different samples, a new SIC paper was used for each surface. All the bonding procedures were performed on the facial and lingual surfaces of mid-coronal dentin.

The samples were divided into 6 groups based on the thickness of the smear layer (thick and thin) on the buccal and lingual aspects and three types of self-adhesive resin cements were used with different pH values. In addition, 96 Valux Plus (3M ESPE, St. Paul, MN, USA) composite resin blocks, measuring 3 mm in diameter and height, were prepared. The chemical compositions and properties of the self-adhesive resin cements used are presented in Table 1.

Group 1: Rely-X Unicem, thick smear layer

The composite resin blocks were bonded to the mid-buccal surfaces of the teeth with the use of Rely-X Unicem (RXU) (3M ESPE, St. Paul, MN, USA) resin cement according to manufacturer’s instructions and then light-cured with Demetron A.2 (Kerr Corporation, 1717 West Collins, Orange, CA 92867) light-curing unit for 10 seconds.

Group 2: Rely-X Unicem, thin smear layer

The composite resin blocks were bonded to the mid-lingual surfaces of the teeth with the use of RXU (3M ESPE, St. Paul, MN, USA) resin cement similar to that in group 1.

Group 3: Clearfil SA Luting, thick smear layer

The composite resin blocks were bonded to the mid-buccal surfaces of the teeth with the use of Clearfil SA Luting (CSL) (Kuraray Noritake Dental INC., Okayama, Japan) resin cement according to manufacturer’s instructions. Light-curing was carried out for 10 seconds.

Group 4: Clearfil SA Luting, thin smear layer

The composite resin blocks were bonded to the mid-lingual surfaces of the teeth with the use of CSL (Kuraray Noritake Dental INC., Okayama, Japan) resin cement similar to group 3.

Table 1: The characteristics of the self-adhesive resin cements used in the present study

| Self-adhesive resin cements | Manufactured by | pH | Composition | Lot number |
|----------------------------|----------------|----|-------------|------------|
| RXU                        | 3M ESPE, St. Paul, MN, USA | <2 | Powder=glass, silica, calcium hydroxide, pigment, substituted, pyrimidine, proxy compound, initiator (filler=72 wt%; average <9.5 um) | 517419 |
| CSL                        | Kuraray, America, New York, USA | 3 | 10-MDP, hydrophobic aromatic dimethacrylate, hydrophobic aliphatic dimethacrylate, colloidal silica, barium glass (filler=66 wt%; 45 vol%; average 2.5 µm) | 0469AB |
| SPC                        | Ivoclar Vivadent, Schaan, Liechtenstein, Germany | 4.5 | dimethacrylate monomers, acid monomers, barium glass fillers, ytterbium trifluoride, silicon dioxide (filler=40 v%; average 5 µm) | T33039 |
**Group 5: Speed CEM, thick smear layer**

The composite resin blocks were bonded to the mid-buccal surfaces of the teeth with the use of Speed CEM (SPC) (Ivoclar Vivadent AG, Bendererstrasse 2, 9494 Schaan, Principality of Liechtenstein) resin cement according to manufacturer’s instructions. Light-curing was carried for 10 seconds.

**Group 6: Clearfil SA Luting, thin smear layer**

The composite resin blocks were bonded to the mid-lingual surfaces of the teeth with the use of SPC (Ivoclar Vivadent AG, Bendererstrasse 2, 9494 Schaan, Principality of Liechtenstein) resin cement similar to group 5.

After 24 hours of storage in distilled water at 37°C, the shear bond strength of the samples were measured in a universal testing machine (Hounsfield Test Equipment, Model HSKS, Surrey, UK) at a strain rate of 0.5 mm/min [Figure 1] and converted to MPa using the following formula: stress = N/mm²

After the shear bond strength test, the fracture modes were determined under a stereomicroscope (Nikon, SMZ800, Tokyo, Japan) at × 40:
- Type 1: Cohesive failure within dentin
- Type 2: Cohesive failure within the composite resin block
- Type 3: Adhesive failure
- Type 4: Mixed failure

**Statistical analysis of data**

Normal distribution of data was evaluated with Kolmogorov–Smirnov test. Data were analyzed with two-way ANOVA. Post hoc least significant difference (LSD) tests were used for two-by-two comparisons of the groups. Statistical significance was set at $P < 0.05$.

**Results**

Table 2 presents the mean bond strength values (MPa) and standard deviations separately for each group.

![Figure 1: Measurement of shear bond strength using Universal Testing Machine](image)

Kolmogorov–Smirnov test showed normal distribution of data ($P > 0.05$). The results of two-way ANOVA showed that the effect of pH of resin cement on the bond strength was significant ($P = 0.02$, $F = 3.74$). However, the differences in bond strength values at different thicknesses of the smear layer were not significant ($P = 0.12$, $F = 2.35$). In addition, the cumulative effect of self-adhesive resin cement pH values and the smear layer thickness was not significant ($P = 0.11$).

Two-by-two comparisons of the bond strength values of the resin cements used with LSD tests showed that the bond strength of RXU resin cement was significantly higher than those of CSL ($P = 0.02$) and SPC ($P = 0.01$) resin cements. However, the difference in the mean bond strength values of CSL and SPC resin cements was not significant ($P = 0.88$). The fracture mode in all the samples was of the adhesive type as evaluated under a stereomicroscope because the dentin was free of any damage and no traces of composite resin and cement were visible on the dentin surface.

**Discussion**

Indirect restorations are the treatment of choice for the restoration of large tooth defects due to their resistance against abrasion and high fracture resistance. One of the factors affecting the durability of indirect restoration is the type and characteristics of the cement used for luting.$^{[1,26]}$ Among the 5 main groups of cements, i.e., zinc phosphate, polycarboxylate, GI, RMGI and composite resin, resin cements are the materials of choice for the cementation of indirect restorations.$^{[1,27]}$ Self-adhesive resin cements are a new generation of resin cements, in which the adhesive and the cement have been incorporated in one step, and there is no need for prior preparation of the tooth surface.$^{[4,5]}$

The results of the present study showed that the RXU self-adhesive resin cement with low pH value had a higher bond strength compared to self-adhesive cements with moderate (CSL) and high pH values (SPC). However, there were no significant differences in the bond strength of self-adhesive resin cements with moderate and high pH values. Similarly, in a study by Mazzitelli et al. the bond strength of RXU was reported to be higher than that of G-Cem with a pH value of 2.7.$^{[28]}$ Furthermore, a study by Barcellos et al. showed that the bond strength of RXU

| Table 2: The mean bond strength values (MPa) and standard deviations of the samples in the present study |
|---------------------------------|--------|-------|
| **Cement** | **Smear layer** | **Mean** | **SD** |
| RXU       | Thick   | 11.88  | 7.041 |
|           | Thin    | 17.22  | 4.88  |
| CSL       | Thick   | 11.18  | 4.00  |
|           | Thin    | 11.26  | 6.35  |
| SPC       | Thick   | 10.98  | 6.58  |
|           | Thin    | 11.03  | 5.44  |

RXU=Rely-X Unicem, CSL=Clearfil SA Luting, SPC=Speed CEM, SD=Standard deviation
and Bifix SE self-adhesive resin cements, both of which have an almost similar pH value <2, were not significantly different.[23] Therefore, it appears differences in pH values and acidity are factors affecting the bond strength of self-adhesive resin cements.

Previous studies have shown that the structural changes in the dentin prepared with different self-etch primers and the depth of penetration of self-etch adhesives into the subsurface dentin depend on the pH of self-etch primer and its ability to decalcify.[21,22] Self-etch adhesives with higher pH values have lower capacity to dissolve the thick smear layer and decalcify the dentin surface compared to self-etch adhesives with low pH.[29]

The difference between adhesive systems and self-adhesive cements in their reaction with tooth structures lies in the fact that in the adhesive systems the water necessary for the release of H+ ions is present in the structure of the adhesive itself; however, in the self-adhesive systems water should be provided by the tooth structure. Although self-adhesive resin cements contain different ingredients in their structure, the bulk of these ingredients consist of methacrylate modified with multifunctional phosphoric acid, which results in self-etching of the tooth without the need for a separate etching step.[23]

During polymerization reaction of the resin cement, the acidic monomers react with water in the tooth structure and ionized in the presence of water by penetration of resin components into the inter-tubular dentin and diffusion through the dentinal tubules. These monomers are neutralized by the calcium in tooth structure during penetration, and the material assumes a neutral pH value.[30] However, Oskoe et al. reported contradictory results.[31] They reported a higher bond strength with the use of a weak self-etching system compared to moderate and strong self-etching systems in relation to their pH; they believed a lack of relationship between decalcification depth and the bond strength of self-etching systems might indicate the role of other factors such as the viscosity of the adhesive, surface tension, and the reaction between the acidic monomers as the water content. Some acidic monomers in the chemical structure of self-etching primers form chemical bonds with hydroxyapatite. Of all the monomers used in the structure of self-etching systems, 10-methacryloyloxydecyl dihydrogen phosphate (10-MDP) has been introduced as a monomer which easily bonds with hydroxyapatite and increases the bond strength. This monomer contains a hydrophobic alky group to create a balance between the hydrophobic and hydrophilic properties of the material and also has double bonds at its ends for more effective polymerization.[29] In the present study, 10-MDP was present in the chemical structure of CSL and given the shear bond strength reported for this cement it can be claimed that the effect of the chemical bond of 10-MDP was not significant to increase the bond strength of this self-adhesive resin cement.

Another significant finding of the present study was the fact that the thickness of the smear layer was not a determinant factor for the shear bond strength of self-adhesive cements to dentin. The smear layer is a noncompact layer of penetrable components with communicating canals. This morphology increases the capacity for diffusion and allows the acidic monomers to gain access to the lowermost layer of the smear layer easily, without considering their aggressive potential. Since the smear layer is a porous layer, the acidic monomers penetrate easily to reach the underlying mineral layer.[29]

Similarly, some previous studies have shown that the performance of the self-etching system has no relationship with the thickness of the smear layer.[18,29,32] Reis et al. attributed the absence of the effect of the thickness of the smear layer on the bond strength of the self-etch bonding system to the morphology of the smear layer and reported that the acidic monomers freely penetrate through the smear layer and reach the underlying mineral layer.[29]

In addition, scanning electron microscope (SEM) observations in a study by Tani and Finger showed that the shear bond strength of all-in-one self-etch adhesives is not influenced by the dentin surface conditions and the thickness of the smear layer does not affect the bond strength. Irrespective of evaluation of the smear layer thickness, they reported that the limitation of their study was the fact that they did not evaluate the quality of the smear layer, which might have affected the penetration of adhesives.[18]

On the contrary, some studies have reported a decrease in the resin-dentin bond strength of self-etching adhesive systems with thicker smear layers.[33,34] Ogata et al. attributed the unfavorable effect of thick smear layers produced by coarse rotary instruments to the occlusion of the orifices of dentinal tubules with smear plugs and reported that the self-etching primer can completely decalcify and eliminate the smear layer, and the bond strength of self-etching primers might be affected by the quantity and quality of the smear layer due to the weak acidity of these primers. Therefore, they suggested that primers be used repeatedly to affect the smear layer to improve the bonding properties.[34]

Similar to De Munck et al., the fracture mode in all the samples was adhesive, irrespective of the cement type and the thickness of the smear layer, which might be attributed to the surface reaction of these cements with dentin and absence of adequate decalcification of dentin and lack of formation of a real hybrid layer on the dentin surface.[15]

SEM evaluations are recommended in future studies to evaluate the quality of the smear layer and its possible effect on the bonding mechanism and also to accurately.
evaluate the bonding of these cements to dentin at different thicknesses of the smear layer.

Conclusion
Considering the limitations of the present in vitro study, it can be concluded that the bond strength of CSL (medium pH) and SPC (high pH) were similar and significantly lower than that of RXU (low pH). However, the smear layer thickness had no significant effect on the shear bond strength of self-adhesive cements.

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Conflicts of interest
There are no conflicts of interest.

References
1. Krämer N, Lohbauer U, Frankenberger R. Adhesive luting of indirect restorations. Am J Dent 2000;13:60D-76D.
2. Carvalho RM, Pegoraro TA, Tay FR, Pegoraro LF, Silva NR, Pasley DH, et al. Adhesive permeability affects coupling of resin cements that utilise self-etching primers to dentine. J Dent 2004;32:55-65.
3. Diaz-Arnold AM, Vargas MA, Haselton DR. Current status of luting agents for fixed prosthodontics. J Prosthodent 1999;8:135-41.
4. Mak YF, Lai SC, Cheung GS, Chan AW, Tay FR, Pasley DH, et al. Micro-tensile bond testing of resin cements to dentin and an indirect resin composite. Dent Mater 2002;18:609-21.
5. Yang B, Ludwig K, Adelung R, Kern M. Micro-tensile bond strength of three luting resins to human regional dentin. Dent Mater 2006;22:45-56.
6. Piwowarczyk A, Bender R, Ottil P, Lauer HC. Long-term bond between dual-polymerizing cementing agents and human hard dental tissue. Dent Mater 2007;23:211-7.
7. Bitter K, Meyer-Lueckel H, Priehn K, Kanjuparambil JP, Neumann K, Kielbassa AM, et al. Effects of luting agent and thermocycling on bond strengths to root canal dentine. Int Endod J 2006;39:809-18.
8. Mazzitelli C, Monticelli F, Osorio R, Casucci A, Toledano M, Ferrari M, et al. Effect of simulated pulpal pressure on self-adhesive cements bonding to dentin. Dent Mater 2008;24:1156-63.
9. Goracci C, Cury AH, Cantoro A, Papacchini F, Tay FR, Ferrari M, et al. Microtensile bond strength and interfacial properties of self-etching and self-adhesive resin cements used to lute composite onlays under different seating forces. J Adhes Dent 2006;8:327-35.
10. Hikita K, Van Meerbeek B, De Munck J, Ikeda T, Van Landuyt K, Maida T, et al. Bonding effectiveness of adhesive luting agents to enamel and dentin. Dent Mater 2007;23:71-80.
30. de Souza Costa CA, Hebling J, Randall RC. Human pulp response to resin cements used to bond inlay restorations. Dent Mater 2006;22:954-62.

31. Oskoei SS, Bahari M, Kimyai S, Navimipour EJ, Firouzmandi M. Shear bond strength of self-etching adhesive systems with different pH values to bleached and/or CPP-ACP-treated enamel. J Adhes Dent 2012;14:447-52.

32. Tay FR, Sano H, Carvalho R, Pashley EL, Pashley DH. An ultrastructural study of the influence of acidity of self-etching primers and smear layer thickness on bonding to intact dentin. J Adhes Dent 2000;2:83-98.

33. Koibuchi H, Yasuda N, Nakabayashi N. Bonding to dentin with a self-etching primer: The effect of smear layers. Dent Mater 2001;17:122-6.

34. Ogata M, Harada N, Yamaguchi S, Nakajima M, Pereira PN, Tagami J, et al. Effects of different burs on dentin bond strengths of self-etching primer bonding systems. Oper Dent 2001;26:375-82.