DR International Dental Research

Original Article

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Bonding performance of different types of expired and unexpired composite resin cements

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Abstract

Aim: The aim of this study was to compare the bond strength of three expired and unexpired composite resin cements used between tooth and ceramic surfaces.

Methodology: In this study, the dentin surfaces of 60 non-carious human teeth and ceramic specimens were bonded with three different resin cements. The resin cements used to bond the teeth and ceramic blocks were divided into two subgroups by expiration date (Group 1a. Variolink II-unexpired, 1b. Variolink II-expired 12 months prior. Group 2a. RelyX Unicem-unexpired, 2b. RelyX Unicem-expired 12 months prior. Group 3a. Panavia F 2.0-unexpired, 3b. Panavia F 2.0-expired 12 months prior). The samples were thermally cycled for 3,000 cycles in a temperature range of 5° C- 55° C. The shear bonding test was performed using a universal tester. The force required for ceramic debonding was recorded. Surface fracture analysis was performed using a stereomicroscope. Mann-Whitney U test was used to compare the bond strength of resin cements with different expiration dates. *P*-value < 0.05 was considered statistically significant.

Results: The difference in bond strength between the unexpired resin and the resin that had expired 12 months before the analysis was found not to be statistically significant in the Variolink II and Panavia pairs but significant in the RelyX resin cement pair.

Conclusion: The self-adhesive resin cements that expired one year earlier showed lower bond strength than their unexpired counterparts; however, the use of expired total-etch and self-etch systems did not significantly affect bond strength.

Keywords: Composite resin cements, expiration date, bond strength, storage conditions



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Received: 4 February 2023 Accepted: 18 May 2023

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How to cite this article:

Karagözoğlu İ, Aksel L. Bonding performance of different types of expired and unexpired composite resin cements. Int Dent Res 2023;13(2):54-59. https://doi.org/10.5577/idr.2023.vol13.no2.2

Introduction

Today, with increasing popularity of full ceramic aesthetic restorations, adhesive cementation has gained more importance. Adhesive cementation is a technique that uses resin cement, requires additional steps on both the prepared tooth surface and the inner surface of the restoration, and requires precision when applied. Composite resin cements used in adhesive cementation are materials with high bond strength, low solubility and high elastic modulus (1,2). Adhesive systems are divided into three groups according to the number of application steps. In a total etch adhesive system, acid, primer and adhesive are applied in separate steps. Then, in order to facilitate this procedure, two-stage adhesives were developed by combining the primer and the bonding. Finally, one-stage (all-in-one) adhesives were produced in which all processes were collected in a single step (3). The ingredients of all dental adhesives are similar. In general. thev contain methacrvlate monomers (hydrophobic and hydrophilic), organic solvents and photoinitiator systems. Common solvents used in dental adhesives are ethanol, acetone, and water. In order to prevent evaporation of dental adhesives, especially acetone-based adhesives must be stored under special conditions (3, 4). Although composite resin cements have many advantages, their chemical and physical structure degrades over time (4). This degradation mainly occurs in two ways: intra-oral degradation that occurs mechanically, chemically and physically and extra-oral degradation that is related to the storage conditions and shelf life of the material (5).

To maintain maximal effectiveness, resin-based materials must be adequately stored. The expiration date or shelf-life describes how long a material retains its maximum properties. Physical and mechanical properties of materials are expected to degrade after expiration. After the expiration date, the polymerization reaction of resin monomers in composite resin cement can be interrupted by the evaporation of organic solvents. Additionally, the permeability and nanoleakage of the hybrid layers can cause problems during bonding (6-8).

Manufacturers generally emphasize that the material should be kept in compliance with the storage conditions, and that it is important to keep the bottles at the appropriate temperature and not to leave the mouth of the bottles open. Most materials used in dentistry degrade rapidly unless the manufacturers' instructions are followed. Composite resin cements, in particular, are sold in sets, so improper storage leads to the deterioration of all the materials they contain. Because these materials are costly, their use after expiration is common.

This study investigated the effects of three different composite resin cements—which expired 12 months before and not yet expired—on the porcelain-dentin bond strength. The null hypothesis tested was that expired composite resin cements had lower bond strengths than unexpired cements.

Materials and Methods

Before commencing this study, the ethics committee of the Gaziantep University Faculty of Dentistry approved it (Desicion no: 2021/393). The study used 60 human molar teeth—which were extracted for periodontal purposes and had no caries and restorations. The crowns and roots of each tooth were separated, and the crowns were embedded in autopolymerized acrylic resin. The teeth's occlusal surfaces were flattened with a lowspeed rotating diamond disc and water cooling. A total of 60 samples in the form of $5 \times 5 \times 2$ mm square prisms were obtained from IPS e.max CAD ceramic blocks. The resin cements used to bond the teeth and ceramic blocks were divided into two subgroups by expiration date (Table 1).

Group 1

- a: Variolink II cement (unexpired)
- b: Variolink II cement (expired 12 months prior)

Group 2

- a: RelyX Unicem cement (unexpired)
- b: RelyX Unicem cement (expired 12 months prior)

Group 3

- a: Panavia F 2.0 cement (unexpired)
- b: Panavia F 2.0 cement (expired 12 months prior)

In group 1, 37% phosphoric acid (Imicryl, Konya, Turkey) was applied to the tooth samples' surfaces for 15 seconds, washed with water, and then air sprayed and dried. Next, Syntac Primer (Ivoclar Vivadent AG, Schaan, Liechtenstein) was applied for 15 seconds, light air was applied for 10 seconds, and Syntac Adhesive (Ivoclar Vivadent AG, Schaan, Liechtenstein) was applied for 10 seconds and spread on the surface with light air. Finally, Heliobond (Ivoclar Vivadent AG, Schaan, Liechtenstein) was applied to the tooth surface for 10 seconds, and air was prayed to spread it over the surface. Then, 9% hydrofluoric acid (Ultradent, South Jordan, Utah, USA) was applied to the ceramic samples. After the hydrofluoric acid was washed with water for 60 seconds and dried with compressed oil-free air, silane (Monobond Plus, Ivoclar Vivadent AG, Schaan, Liechtenstein) was applied for 60 seconds and dried. Then, Variolink Base and Variolink Catalyst Low Viscosity at a 1:1 ratio were mixed for 10 seconds with a spatula and applied to the tooth surface. Finally, the prepared ceramic samples were cemented to the teeth.

In group 2, 9% hydrofluoric acid was applied to the ceramic samples, and then they were washed and dried. The cement capsule was placed in the system activator and opened by holding down the activator handle for 2-4 seconds. After mixing, the capsule was placed in its applicator. The cement was evenly distributed over the tooth surface. Ceramic specimens were placed on the tooth surface using finger pressure.

Table 1. Chemical compositions of the resin cements used in the study.

Material	Manufacturer	Composition			
Variolink II	Ivoclar- Vivadent, Shaan, Liechtenstein	Monomer matrix: Bisphenol A diglycidyl methacrylate (Bis-GMA), urethane dimethacrylate (UDMA), triethylene glycol dimethacrylate (TEGDMA). Inorganic load: barium glass, ytterbium trifluoride, barium fluorsilicate glass and aluminum, mixed spheroidal oxides (particle size: 0.04-3 µm; mean filler size: 0.7 µm; base: 46.7 vol%, low viscosity catalyst: 43.6 vol%).			
Rely-X Unicem	3M ESPE, St. Paul, MN, USA	Base paste: methacrylate monomers containing phosphoric acid groups, methacrylate monomers, silanated fillers, initiator components, stabilizers. Catalyst paste: methacrylate monomers, alkaline (basic) fillers, silanated fillers, initiator components, stabilizers, pigments.			
Panavia F 2.0	Kuraray, Okayama, Japan	Catalyst paste: Bis-GMA, TEGDMA, glass particles A paste: silanized silica particle, silanized colloidal silica, MDP, hydrophilic aliphatic D, aromatic hydrophilic D, camphorquinone, catalyst, initiator B paste: silanized barium glass, sodium fluoride, aromatic hydrophilic D, aliphatic hydrophilic D, pigment catalysts (load content ≅ 76%)			

In group 3—as in the other groups—9% hydrofluoric acid was applied to the ceramic samples, and they were washed and dried. Panavia F 2.0 ED Primer liquid A and liquid B were mixed in equal amounts in their own mixing bowls. The mixture was applied to the tooth surface using a disposable brush and left for 30 seconds. Panavia F 2.0 Pastes A and B were mixed in equal amounts on mixing paper with a plastic spatula for 20 seconds and placed on the tooth surface. Then, the ceramic samples were placed on the tooth surface using finger pressure and cured with an LED light device.

All three groups were kept in distilled water at 37° C for 24 hours and placed in a thermal cycler (SD Mechatronic GMBH, Feldkirchen-Westerham, Germany). The samples were thermally cycled for 3,000 cycles in a temperature range of 5° C- 55° C. The shear bonding test was performed using a universal tester (Shimadzu AG-XD 50 kN, Shimadzu Corporation, Kyoto, Japan) at a speed of 1 mm/min. Shear force was applied using a one-sided cutting blade parallel to the tooth and ceramic interface. The force required for ceramic debonding was recorded in Newtons (N) and converted to megapascals (MPa). Surface fracture analysis was performed using a stereomicroscope.

Statistical analysis

Analyses were performed by using SPSS software (IBM SPSS Statistics version 24.0, IBM Inc., Armonk, NY, USA).

Due to the data's non-parametric distribution, the Mann-Whitney U test was used to compare the bond strength of resin cements with different expiration dates. P-value < 0.05 was considered statistically significant.

Results

The shear bond strength values of the tested groups were statistically analyzed. The standard deviation and mean values of the shear bond strength results for the Variolink II, RelyX Unicem, and Panavia F 2.0 cements are summarized in Table 2 by expiration date.

According to the statistical analysis results, when the expired groups were compared to each other in terms of shear bond strength value, a significant difference was found between the Variolink II and RelyX Unicem resin cements (p = 0.041), but there was no statistically significant difference between the Variolink II and Panavia F 2.0 groups or the Panavia F 2.0 and RelyX Unicem groups. There was no statistically significant difference between the unexpired and expired cements before the analysis in the Variolink II and Panavia F 2.0 groups (p = 0.258 and p = 0.083, respectively). However, a significant difference was found between the unexpired and expired RelyX Unicem resin cement (p =0.006). When unexpired cement groups were compared to each other, there were no statistically significant differences (p > 0.05).

 Table 2. Descriptive statistics for the shear bond strength values.

	ι	Jnexpired	Expired		
	Mean MPa	Standard Deviation	Mean MPa	Standard Deviation	
Variolink II	19.08	5.44	16.1	5.02	
RelyX Unicem	16.63	4.15	10.99	4.47	
Panavia F 2.0	19.98	5.69	14.65	5.87	

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The fracture surfaces of the samples in all groups, obtained as results of the shear bond strength test, were examined under a stereomicroscope at 20x magnification. Types of failure were classified as follows: A: cohesive on dentin surface, B: adhesive on cementtooth interface, C: adhesive-cohesive on tooth surface, D: cohesive in cement, E: adhesive-cohesive in ceramic (mix), and F: adhesive on ceramic-cement interface. The detected failure types are given in Table 3. Adhesive fracture was observed at the cement-tooth interface in 65% of all samples; 28% showed cohesive fracture in the cement itself, 2% showed cohesive fracture within the tooth, and 5% showed both adhesive and cohesive fractures on the tooth surface.

Table 3. Failur	e types in	luting cement	and specimens	by group.
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Group		Ν	Failure type					
			Α	В	С	D	E	F
Varializk II	Unexpired	10	-	8	2	-	-	-
	Expired	10	-	7	1	2	-	-
Poly V Unicom	Unexpired	10	1	5	-	4	-	-
Kety-A Unicem	Expired	10	-	5	-	5	-	-
	Unexpired	10	-	7	-	3	-	-
Fallavia F 2.0	Expired	10	-	7	-	3	-	-

Discussion

In this study, the bond strengths of Rely-X Unicem, a selfadhesive cement that is easily applied; Panavia F 2.0, a self-etch system that is relatively easily applied; and Variolink II, a total-etch system, were compared according to different expiration dates. All of these materials were applied with an adhesive cementation technique. The shelf life and storage conditions of the adhesive or bonding applied with the adhesive cementation technique are very important. Because of the decomposition of the additives in the adhesives (initiators/stabilizers), the evaporation of the components and the polymerization or hydrolysis of the monomers occur over time (9). The shelf-life of composite resin cement materials used in dental clinical practice is critical. Restoration failures may be caused not just by inadequate clinical practices, but also by the limited shelf life of certain of the materials used. Previous studies examining the shelf life of dental bonding and adhesive materials have concluded that the use of the universal adhesive system beyond the expiry limit results in decreased bonding performance (10-13). However, the examination of the effect of bonding or adhesive alone is insufficient because adhesive systems provide tooth-restoration bonding as a whole. Therefore, in this study, adhesive, bond, and composite resin were evaluated as an adhesive system.

The solvent type of the primer incorporated into the adhesive structure is an important factor in terms of dentin bond strength. Adhesives systems with water and ethanol based-primer, contain hydrophobic dimethacrylates in their structure. Dimethacrylates are not water resistant, over time they hydrolyze into methacrylic acid and cross-couplings in the bottle. Studies have determined that this disintegration reduces

and self-etching systems had not a significant impact on bond strength. This difference can be explained by differences in the structure of the adhesive used, the monomer content, and the solvent used in the primer construction.
Many studies have examined the mechanical and physical properties of composite resins or resin-based materials according to their shelf life (14-16). In these studies, mechanical properties such as microhardness, water solubility, degree of conversion, static modulus of

the bond strength by disrupting the structure of the

adhesive over time (12, 13). In the current study, the 12month expired self-adhesive composite cement had

lower bond strength, but the use of expired total etching

water solubility, degree of conversion, static modulus of elasticity, and flexural strength of expired and nonexpired composite resins were investigated. In some studies on cements, the mechanical properties of expired and non-expired cements have been compared (17, 18). Wajong et al. investigated the effects of shelf life on the compressive strength of resin-modified glass ionomer cement and concluded that there was a decrease in the compressive strength value according to expiration date (18). In their study on glass ionomer cements, Alonso et al. reported that the microhardness value decreased as the expiration date approached (17). There is no study in the literature investigating the bond strength of expired and non-expired resin cements with dentin. Thus, in the present study, the bond strengths of three different resin cements were investigated according to whether they have expired.

In general, a temperature between 4°C and 20°C is advised. However, as the storage conditions of resin composites can vary depending on the geographic and climatic conditions (sun exposure and humidity) of the country, the majority of dental manufacturers advise storing resin composites in the refrigerator (8). Some clinicians store resin composites in refrigerators at 2-5 °C in order to extend their expiration life, particularly in regions with a hot climate (19). Ma et al. reported that storage conditions affect the clinical performance of resin materials, especially self-etch resins (20). Previous studies have reported that self-etch adhesive systems have lower bonding performance than total-etch adhesives (21, 22). However, other studies have reported that self-etch and total-etch systems have similar bonding performances (23, 24). In the present study, when the total-etch system (Variolink II) was compared to the self-etch system (Panavia F 2.0), the bond strength values were similar. However, the bond strength was lower in the self-etch system than in the total-etch system in the expired group, but the difference was not statistically significant.

Various studies have reported on the bonding performance of self-adhesive resin cements on dentin. For example, Rely-X Unicem (self-adhesive system) has a similar bond strength to conventional resin cements (25, 26). Viotti et al. investigated the bond strengths of conventional resin cements and self-adhesive resin cements (Rely-X Unicem, Maxcem, and G-cem) on dentin by applying a microtensile test (23). They reported that self-adhesive resin cements exhibited significantly lower bond strengths than some of the conventional resin cements (23). In our study, similar to Viotti et al.'s results, the bond strength of Rely-X Unicem was found to be lower than those of the other tested cements. In addition, the bond strength of the expired self-adhesive resin cement was significantly lower than those of the other expired cements.

Taking into account the storage conditions and duration of the one-step self-etching adhesives, similar to the results of the present study, it was reported that a decline in adhesive strength values was observed at the end of the storage period (27).

the present study, according the In to stereomicroscopic examination of the fractured surfaces, adhesive type failure was observed at the dentin-cement interface in 65% of all samples, while fracture within the cement itself was detected in 28%. The failure rate of the cement was similar to the bond strength results. The bond strength was lowest and the failure rate was highest in the expired self-adhesive (Rely-X Unicem) group. No failure was detected at the cement-ceramic interface in any group. In other words, resin cements showed a strong bond to the IPS e.max ceramic surface roughened with hydrofluoric acid, but they did not exhibit similar bonding to the dentin surface.

Conclusion

For Variolink II and Panavia F 2.0, the bond strengths of resin cements that were used after their expiration past one year were similar to those of non-expired cements. When Rely-X Unicem was used one year after its expiration date, the bond strength values were significantly lower than those of the unexpired cements. This difference between the groups may be due to the difference in the adhesive cementation technique.

Resin cements stored in appropriate conditions according to manufacturer instructions can be used in clinical conditions when evaluated for bond strength; however, there is a need for more studies examining the mechanical, physical, and biological effects of expired cement.

Disclosures

Ethical Approval: Ethics committee approval was received for this study from Gaziantep University Dental Ethics Committee, in accordance with the World Medical Association Declaration of Helsinki, with the approval number: 2021/393).

Peer-review: Externally peer-reviewed.

Author Contributions: Conception - I.K.; Design - I.K., L.A.; Supervision - L.A.; Materials - I.K., L.A.; Data Collection and/or Processing - I.K.; Analysis and/or Interpretation - I.K., L.A.; Literature Review - I.K., L.A.; Writer -I.K.; Critical Review -L.A.

Conflict of Interest: No conflict of interest was declared by the authors.

Funding: The authors declared that this study has received no financial support.

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