Does Adhesive Resin Application Contribute to Resin Bond Durability on Etched and Silanized Feldspathic Ceramic?

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Purpose: To assess the effect of adhesive application and aging on the bond durability of resin cement to etched and silanized feldspathic ceramic.

Materials and Methods: Twenty blocks (6.4 x 6.4 x 4.8 mm) of feldspathic ceramic (Vita VM7) were produced. The ceramic surfaces were conditioned with 10% hydrofluoric acid gel for 60 s and silanized. They were then randomly divided into two groups. While half of the group received no adhesive, in the other half, a layer of adhesive (Scotchbond Multi-Purpose Plus) was applied. Each ceramic block was then placed in its silicone mold with the treated surface exposed. The dual-cured resin cement (Variolink II) was injected into the mold over the treated surface and polymerized. Specimens were sectioned to achieve nontrimmed bar specimens (approximately 12 sticks/block) that were randomly divided into 2 groups: a) non-aged – microtensile bond test immediately after sectioning; b) aged-thermocycling (TC) 12,000 times, 5°C to 55°C, and water storage (50 days). The microtensile bond strength test was performed in a universal testing machine (crosshead speed: 1 mm/min). The failure types were examined using an optical light microscope and SEM. Bond strength results were analyzed using two-way ANOVA and Tukey's test ($\alpha = 0.05$).

Results: The adhesive application affected the bond strength results significantly ($p = 0.0001$) (without adhesive > with adhesive). While aging conditions did not reduce the bond strength in the groups that received no adhesive (20 ± 5.3 MPa non-aged and 21.5 ± 5.6 aged) ($p = 0.1698$), it significantly affected the bond strength results of the group with adhesive application (18 ± 4.4 MPa to 14.4 ± 4.7 MPa) ($p < 0.001$). All groups showed mainly mixed type of failures between the ceramic and the resin cement (81% to 100%). The group in which no adhesive was applied presented a higher incidence of cohesive failure of ceramic after aging (18%) than those of the other groups.

Conclusion: The use of adhesive did not improve resin cement adhesion to the etched and silanized feldspathic ceramic after long-term thermocycling and water storage.

Keywords: acid etching, bond durability, glass ceramic, microtensile test, resin cement.

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For durable adhesion of resin-based materials in the repair of fixed dental prostheses (FDP) or adhesive cementation of glass-ceramic restorations, the standard procedure for conditioning the surfaces of such ceramics involve etching with hydrofluoric acid (HF) gel, silanization, and subsequent intermediate adhesive resin (hereafter termed "adhesive") application. Acid-sensitive, glass-based ceramics such as feldspathic, leucite, and lithium disilicate ceramics undergo surface degradation by HF acid gel, yielding a topographic pattern that favors micromechanical bonding. In addition, due to its bifunctional characteristics, application of silane coupling agent on the etched ceramic surface increases the chemical adhesion between the ceramic and resin materials.[1,5,12] This process promotes the cement wettability on the ceramic surface,[11,14] enhancing the contact with resin cements. Moreover, silane coupling agents couple the silica (silicon oxides) present in glassy-matrix ceramics to the organic matrix of resin cements by means of siloxane bonds.[15,19] Application of a hydrophobic adhesive layer after silane application is frequently advised by the manufacturers, since it increases the wettability especially of bis-GMA-based resin materials, due to their lower viscosity compared to resin cement.

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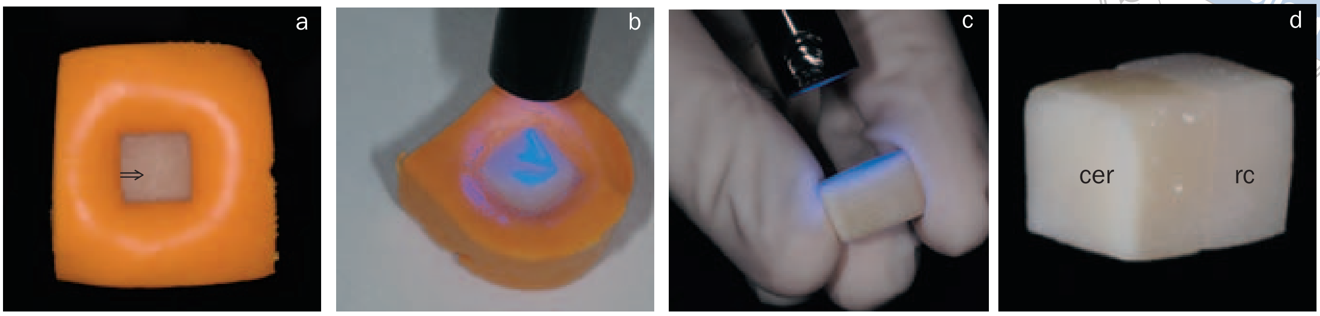

Fig 1 (a) The ceramic block was pushed inside the silicone with the conditioned surface free (c) to receive the resin cement; (b) resin cement was applied to the ceramic surface and photoactivated; (c) the ceramic/cement set was removed from the silicone and the cement was photoactivated again; (d) ceramic/cement assembly (cer= ceramic; rc= resin cement).

While some studies suggest that chemical adhesion promoted by a silane coupling agent is the major mechanism responsible for adhesion of resin-ceramic systems and is crucial for durable bond strength,[2-4,8,16,18] others reported that etching makes the greatest contribution to the final bond strength.[17,18] According to some theories, adhesives serve as shock-absorbing layers between the resin/resin assemblies.[10] The additional function of adhesive remains controversial,[13,16] even among the same study groups.[6,7] In these studies, either different testing methods were used or the specimens were not subjected to long-term aging conditions.[5,6,13,16]

Therefore, the objective of this study was to assess the effect of adhesive application and aging on the bond durability of resin cement to etched and silanized feldspathic ceramic.

MATERIALS AND METHODS

Twenty blocks (6.4 x 6.4 x 4.8 mm) of feldspathic ceramic (VITA VM7, Dentin 5M2, Vita Zanhfabrik; Bad Säckingen, Germany; batch #7404) were produced according to the manufacturer's instructions. The cementation surface of each ceramic block was leveled and polished in a machine using silicon carbide papers (3M; St Paul, MN, USA) in sequence (600-, 800-, and 1200-grit) under water cooling. Impressions were made from each ceramic block with addition silicone putty (Elite HD, Zhermach; Badia Polesine, Italy; batch #18443). The ceramic block was pushed inside the silicone in order to achieve 3 mm distance between the upper portion of the mold and the surface of the block to create space for the resin cement (Fig 1).

Prior to surface conditioning, all blocks were ultrasonically cleaned (Vitasonic, Vita Zanhfabrik) for 5 min using distilled water. The ceramic surfaces were etched with 10% HF acid gel (Dentsply; Petropolis, Brazil; batch #235100) for 60 s, rinsed with air-water spray for 60 s, and air dried. The ceramics were cleaned ultrasonically in distilled water for 4 min. Then one layer of a silane coupling agent was applied (Porcelain Primer, Bisco; Schaumburg, IL, USA; batch #04000110696) with a clean brush and left to react for 5 min.

The ceramic blocks were then randomly divided into two groups (n = 10 blocks per group). While half of the ceramic blocks received no adhesive, in the other half, a layer of adhesive (Scotchbond Multi-Purpose Plus, 3M ESPE; Seefeld, Germany; batch #5NY) was applied according to the manufacturer's instructions. Each ceramic block was then placed in its silicone mold with the treated surface exposed. The dual-curing resin cement (Variolink II, Ivoclar-Vivadent; Schaan, Liechtenstein; batch #G26358) was mixed following the manufacturer's instructions and injected into the mold on the treated surface of the ceramic block using a centrix syringe (DFL; Rio de Janeiro, Brazil). The cement in the mold was photoactivated (XL 3000, 3M ESPE; St Paul, MN, USA) with a light output of 500 mW/cm^2 for 40 s on each side of the specimen. The intensity of the light was verified by a radiometer to not be lower than 500 mW/cm^2 (Demetron LC, Kerr; Orange, CA, USA) before starting the polymerization in each group. Oxygen inhibiting gel (Oxyguard II, Kuraray; Okayama, Japan; batch #00482A) was applied on the free surfaces. After 10 min, the ceramic-block/resin-cement assembly was removed from the mold and the cement was once again submitted to light polymerization from the five aspects of the block (upper and lateral) for 40 s per side.[10]

Specimen Preparation for the Microtensile Bond Strength Test (μTBS)

Ceramic/cement blocks were sectioned using a diamond disk (Microdont; São Paulo, Brazil, no. 34570) at low speed under water cooling in a sectioning machine (Lab-Cut 1010, Extec; Enfield, CT, USA). Initially, the cemented blocks were fixated with cyanoacrylate adhesive gel (Super Bonder Gel, Loctite; São Paulo, Brazil) on a metallic base that was attached to the sectioning machine. The blocks were positioned as perpendicularly as possible in relation to the diamond disk of the machine. The first section, measuring approximately 1 mm, was discarded to eliminate the possibility of excess or absence of cement at the interface that might alter the results. Thereafter, two sections measuring 1.0 ± 0.1 mm in thickness were achieved. Each section was rotated 90 degrees and once again fixated to the metallic base. The first section was discarded (1 ± 0.1 mm) for the previously mentioned reasons. Subsequently, four other sections were achieved, also measuring 1.0 ± 0.1 mm in thickness. This process was followed for the other two sections, and thus only the central specimens were used for the experiments.[9] It was planned to obtain approximately 12 specimens from each block. The beam specimens had nonmachined (nontrimmed) bonding areas with a bonded area measuring approximately 1.0 ± 0.1 mm^2 and 8 mm length.

The bar specimens obtained from each ceramic block were randomly divided into 2 testing conditions (n = 50). For the non-aged group, after sectioning, the specimens were immediately submitted to the microtensile test. For the aged group, specimens were submitted to thermal cycling (12,000 cycles; 5°C to 55°C, dwell time: 30 s, transfer time: 2 s) (Nova Etica; São Paulo, Brazil) and stored in distilled water at 37°C for 50 days before microtensile testing was performed. Thus, 4 groups were obtained, considering the "surface treatment" (2 types) and "storage condition" (2 types) (Table 1).

Table 1 Test groups considering adhesive resin application and aging conditions

Adhesive	Storage condition	Group (n = 50)
Without	Non-aging*	1
With	Non-aging*	2
Without	Aging**	3
With	Aging**	4

*microtensile test immediately after cutting; **thermocyling 12,000 times; 5°C to 55°C; water storage in distilled water at 37°C for 50 days.

Microtensile Bond Strength Test

Keeping the adhesive zone free, each specimen was affixed with cyanoacrylate gel (Super Bonder Gel, Loctite) to the rods of an alignment device adapted for this test. The specimens were positioned parallel to the long axis of the device in order to reduce the bending stresses. The device was fixated in the universal testing machine (EMIC DL-1000, EMIC; São José dos Pinhais, Brazil) as parallel as possible to the application of the tensile load, and testing was performed at a crosshead speed of 1 mm/min.

The bond strength was calculated according to the formula R=F/A, where "R" is the strength (MPa), "F" is the load required for rupture of the specimen (N), and "A" is the interface area of the specimen (in mm^2), measured with a digital caliper before the test.

Failure Type Analysis

All specimens (N = 200) submitted to the microtensile test were analyzed using an optical light microscope (MP 320, Carl Zeiss; Jena, Germany) at 100X to 5000X magnification, and some representative specimens were selected for scanning electron microscopy (SEM) (JEOL JSM T330A, JEOL; Tokyo, Japan) at 75X and 200X magnification to observe the type of failure. Failures were classified as follows: adhesive between ceramic and cement (ADHES); cohesive failure of the cement (COHES-cem); cohesive failure of cement and ceramic (MIX).

Statistical Analysis

The microtensile data were analyzed by two-way ANOVA, with bond strength as the dependent variable, and the adhesive application and the storage conditions as the independent factors (Statistix 8.0 for Windows, Analytical Software; Tallahassee, FL, USA). P-values less than 0.05 were considered to be statistically significant in all tests. The beam was used as the experimental unit, since the aging affect was tested on the beams instead of the blocks.

RESULTS

Two-way ANOVA revealed a significant influence of the adhesive application (p = 0.0001), but aging conditions did not show a significant influence (p = 0.1698) on the bond strength results. Interaction between factors was also significant (p = 0.0004) (Tukey's test) (Table 2).

Table 2 Results of two-way ANOVA for the experimental conditions

Source	DF	SS	MS	F value	p-value*
Adhesive application	1	1045.71	1045.71	41.58	0.0001
Storage/thermocycling	1	47.75	47.75	1.90	0.1698
Interaction	1	328.96	328.96	13.08	0.0004
Residual	196	4929.14	25.15		
Total	199	6351.55			

*p < 0.05

Table 3 **Mean values and standard deviations of the μTBS results (MPa) per group**

Adhesive	Non-aging	Aging
Without	20.0 ± 5.3^{ab}	21.5 ± 5.6^{a}
With	18.0 ± 4.4^{b}	14.4 ± 4.7^{c}

*The same superscript letters indicate no significant differences (p > 0.05).

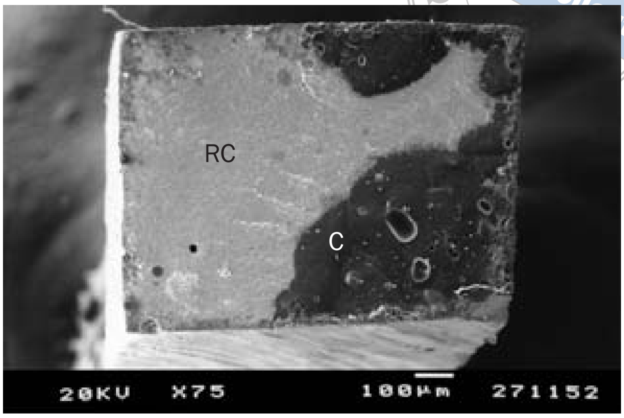

Fig 2a Representative micrograph of the surface of a debonded specimen from group 4 with mainly cohesive failure of cement (COHES-cem) (75X) (RC = resin cement).

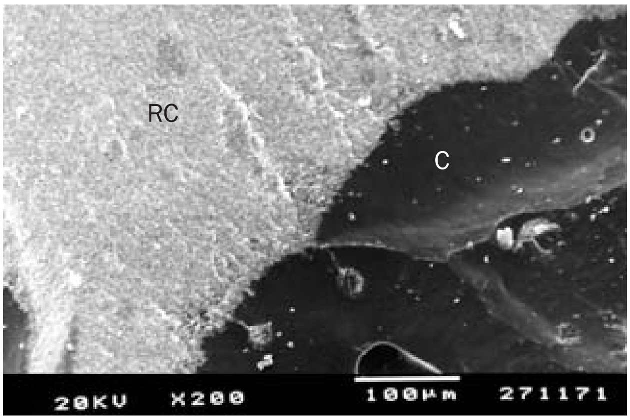

Fig 2b Representative micrograph of the surface of a debonded specimen from group 2 showing cohesive failure of cement and ceramic (MIX) failure (75X) (C = ceramic, RC = resin cement).

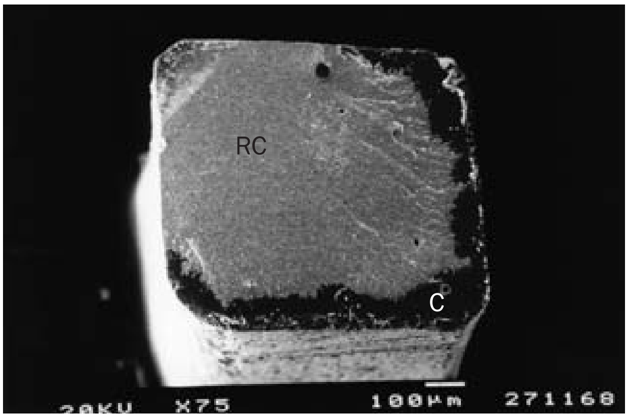

Fig 2c Micrograph of the surface of the specimen in Fig 2b showing cohesive failure of ceramic 200X (C = ceramic, RC = resin cement).

Table 4 **Number of tested specimens per group and incidence of failure types in percentage (%) after the microtensile test per experimental group**

Group	Total no. (%) of specimens	ADHES	Failure type* COHES-cem	MIX
1	50 (100%)	0	1 (2%)	49 (98%)
2	50 (100%)	0	0 (0%)	50 (100%)
3	50 (100%)	0	9 (18%)	41 (81%)
4	50 (100%)	0	4 (8%)	46 (92%)
Total	200 (100%)	0	14 (7%)	186 (93%)

Failure between ceramic and cement (ADHES); cohesive failure of cement and ceramic (MIX); cohesive failure of the cement (COHES-cem).

The mean bond strength decreased significantly from 18 ± 4.4 MPa to 14.4 ± 4.7 MPa after 12,000 thermal cycles and 50-day water storage (p = 0.0001) in the group in which adhesive was applied. Aging conditions did not reduce the bond strength in the groups that did not receive adhesive (20 ± 5.3 MPa non-aged, and 21.5 ± 5.6 MPa after TC and water storage) (p > 0.05) (Table 3).

Failure analysis demonstrated that all groups showed mainly MIX failures between the ceramic and the resin cement (81% to 100%). The group in which no adhesive was applied presented an increased incidence of cohesive failure of resin cement (COHES-cem) after aging (18%) (Table 4).

SEM micrographs representing the failure types of the debonded specimens are presented in Fig 2.

DISCUSSION

For durable adhesion of resin-based materials to glassy matrix ceramics, the sequence of surface conditioning prior to resin adhesion is of utmost importance. The necessity of HF etching and silanization has been well established and is an accepted procedure for conditioning feldspathic ceramics. However, the use of adhesive has been reported to be optional in several studies.[2,6,7,16]

In the study by El Zohairy et al,[7] which also used microtensile bond strength tests, adhesives that contain hydrophilic monomers were found to have a negative influence on the resin-ceramic bond durability. However, in that study, the block ceramic/resin specimens were aged for only 1, 7, and 28 days. After 28-day water storage, the bond remained stable when hydrophobic bonding was used. Similarly, Peumans et al,[13] employing a microtensile bond strength test setup, found that the use of adhesive did not yield significant differences compared to the groups where no adhesive was used, when bonding Variolink II to the leucite-reinforced CAD/CAM ceramic PROCAD. Yet they, too, performed the bond strength tests after only 24-h water storage. They reported that some specimens failed before actual testing, and the mictotensile was determined from the specimens that survived specimen processing. Nonetheless, the number of excluded specimens was not mentioned. Failure types were reported to be 48% mixed. Spohr et al[20] also studied the resin cement adhesion to glass ceramic using the tensile test, and the results were more favorable with HF and silanization only. Again, this study used only 24-h water storage at 37°C; therefore, no aging effect could be extrapolated from this study.

The results of this study showed significant decrease in bond strength (from 18 ± 4.4 MPa to 14.4 ± 4.7 MPa) after 12,000 thermal cycles and 50 days of water storage in the group where adhesive was applied. On the other hand, aging conditions did not decrease the bond strength in the groups that did not receive adhesive (20 ± 5.3 MPa and 21.5 ± 5.6 MPa for non-aging and aging conditions, respectively).

The aging duration necessary to represent the worst-case scenario for resin-ceramic adhesion studies is not clear in the dental literature, but it is known that water storage and thermocycling results in hydrolytic degradation of the ceramic/resin interface.[17] Resin-based materials absorb water to a certain time-dependent degree during water storage, taking days or weeks to reach maximum absorption. It should also be noted that in this study, aging procedures were applied to the bars (sticks), not to the blocks. The reason for this was to simulate margins of restorations that are directly exposed to the aging conditions rather than the inner portions of the adhesive interfaces. One can anticipate that the water sorption to the inner parts of the blocks would take longer than the water sorption of the bars of approximately 1.0 ± 0.1 mm^2 adhesive surface area.

In the studies by Reich et al[16] after 10,000 thermocycles, and Blatz et al[3] after 12,000 thermocycles and 180-day water storage, adhesion of Variolink II to feldspathic ceramics did not show significant differences with and without adhesive. However, both studies were conducted using shear testing, resulting in mainly cohesive failures of the substrates. Cohesive failures experienced due to the nature of the shear test have been demonstrated earlier.[4] The analysis of the debonded surfaces under an optical light microscope and SEM demonstrated mainly mixed failures between the ceramic and the resin cement.

In terms of composition, adhesives exist either in hydrophobic or hydrophilic forms. The adhesive in this study was hydrophobic, containing HEMA and bis-GMA and not a solvent. Adhesives with solvent presented more water absorption, solubility, and a greater diffusion coefficient than the adhesives without solvent.[8] Water absorption is influenced by the affinity of the material to water, which also depends on the amount of hydroxyl groups (OH) in the resin matrix, creating hydrogen links with the water.[8] The adhesive tested in this study contains bis-GMA and HEMA, where the latter is hydrophilic, favoring water absorption. Foxton et al[9] noted that the evaporation of the solvent in the silane may increase the number of siloxane bonds to the ceramic surface, making the adhesion more resistant to hydrolytic degradation when immersed in water for a long period of time.

Thus, taking the results of this current study into account and from the clinical point of view, the applicaton of adhesive on the HF-etched and silanized feldpathic ceramic may not be necessary. The decrease of 5 MPa may not be relevant for the retention of the adhesively cemented FDPs, and therefore adhesive application could still be considered “optional”. However, it may affect the marginal degradation or discoloration of the margins, since the application of adhesive adds to the thickness of the adhesive interface.

CONCLUSION

The use of an intermediate adhesive resin did not improve resin cement adhesion to the etched and silanized feldspathic ceramic after long-term thermocycling and water storage.

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Clinical relevance: During repair or cementation of feldspathic-ceramic–based restorations using resins containing bis-GMA, the application of a HEMA-containing hydrophilic intermediate adhesive resin does not seem to be necessary.