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Impact of Silane-containing Universal Adhesive on the Biaxial Flexural Strength of a Resin Cement/Glass-ceramic System

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Clinical Relevance

Using an efficient silane primer is crucial to improve an indirect bonded ceramic restoration's mechanical properties and to preserve interface integrity with resin cement when submitted to load.

SUMMARY

The aim of this study was to determine whether using a silane-containing universal adhesive as a silane primer in glass-ceramic/resin cement systems affects biaxial flexural strength (BFS) and bonded interface integrity after loading. Glass-ceramic (IPS e.max CAD, Ivoclar/Vivadent, Schaan, Liechtenstein) disc-shaped specimens (6.5±0.1mm in diameter, 0.5±0.1mm thick) were etched with 5% hydrofluoric acid (HF) for 20 seconds and divided into four groups of 30 specimens, to be treated as follows: 1) One bottle silane primer (RCP); 2) Separate application of silane and adhesive (RCP+SB); 3) Silane-containing universal adhesive (SBU); 4) No treatment (C). After silanization, all specimens were resin cement– coated and polymerized for 40 seconds. Each specimen layer was measured, as well as each assembly's thickness, using a digital caliper and scanning electron microscope (SEM). Specimens were stored for 24 hours and submitted to a BFS test (1.27 mm/min). BFS values were calculated using the bilayer disc-specimen solution. Bonded interfaces were analyzed on fractured fragments using SEM. One-way ANOVA and Tukey tests (α=0.05) were applied, as well as the Weibull analysis. Factor "silane treatment" was statistically significant (*p*<0.0001). RCP+SB (372.2±29.4 MPa) and RCP (364.2±29.5 MPa) produced significantly higher BFS than did the C (320.7±36.3 MPa) or SBU (338.0±27.1 MPa) groups. No differences were found in the Weibull modulus (*m*: RCP: 10.1-17.3; RCP+SB: 10.1-17.0; SBU: 12.3-22.4; C: 7.4-

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12.9). Bonded interface analysis exhibited ceramic-cement separation (SBU, C) and voids within the resin cement layer (all groups). Neither the ceramic/cement system's BFS nor its bonded interface stability were improved by SBU after loading.

INTRODUCTION

Metal-free restorations are a suitable clinical option for indirect procedures because of their biocompatibility, excellent mechanical properties, and optimal esthetics.[1] In view of these properties, lithium disilicate glass-ceramic can be considered the material of choice for many clinical situations. To achieve a proper bonding between inorganic restorative materials and organic tooth tissues, coupling agents capable of linking both surfaces must be applied to them.[2-4] Moreover, the use of resin cement to bond ceramic restorations to teeth may improve the mechanical performance at the tooth-ceramic junction.[5,6] To ensure long-lasting tooth-restoration bonding, all these issues must be considered.[7]

Glass-ceramic surface treatment must be performed in order to enhance resin cement/glass-ceramic bonding. Two main approaches are recommended in the literature: mechanical and chemical.[2] The mechanical strategy consists of etching the ceramic surface using hydrofluoric acid (HF), which produces a selective glass-content removal, exposes the crystalline structure, raises surface energy, and facilitates mechanical interlocking of the resin cement.[4,5,8,9] Otherwise, silane couplers provide chemical adhesion between resin cements and silica-containing ceramic substrates.[5] Silane molecules employed in dentistry contain two functional groups: one reacting with polymerizable methacrylates, the other reactive toward silica in glassy structures. The alkoxy groups of this molecule must be activated by a hydrolyzation process (SiOR→SiOH), being suitable then to undergo a condensation reaction when in contact with a ceramic surface, in which water is released as a byproduct.[2] The methacrylate group reacts with the polymerizable side of the resin cement[2] to achieve a three-dimensional, cross-linked network between the ceramic and resin cement.[10-12] This chemical process, complemented with mechanical interlocking, is currently the most accepted procedure for enhancing resin cement/glass-ceramic bonding.[2,4,5]

Following a simpler approach for dental bonding, manufacturers have also added silane to dental adhesives, specifically to a category of materials known as "universal adhesives." These normally contain phosphate acid monomers, such as 10-methacryloyloxydecyl dihydrogen phosphate (MDP), which can chemically bond with hydroxyapatite (making it possible to eliminate dentin etching with phosphoric acid), metallic ions present in some ceramics (mainly polycrystalline ceramics), and methacrylate groups of resin cements.[13-15] The addition of silane makes a chemical interaction with glass-containing ceramics possible, amplifying the range of substrates in which these adhesives will act as bonding promoters for resin-based materials.[16] Derived from this, manufacturers claim that silane-containing universal adhesives can be used as conventional silane primers. However, some studies have reported that universal adhesives produced lower ceramic-cement bond strength than conventional silane primers or separate use of silane and adhesive,[17-19] probably because of some kind of silane inactivation inside universal adhesives of low pH or other reason.[19-20]

This broad range of surface treatments, showing different effectiveness, may not only affect glass-ceramic/resin cement adhesion, but its mechanical properties as well. Related to this, the influence of some surface treatment protocols on a material's mechanical performance has also been evaluated. Previous studies have shown an increase in ceramic flexural strength when applying an adhesive, unfilled resin coat,[21-24] or resin cement[12] after a silane coupler. Conversely, another investigation found that the application of conventional silane alone (not resin-cement coated), exerted no effect on ceramic biaxial flexural strength, being more involved with the ceramic's surface texture and unfilled resin application.[25] However, the effect of different types of silane primers (probably showing dissimilar bonding-promoting effectiveness between them) on the flexural strength of ceramic/cement systems is still uncertain.

Thus, the way luting procedures are managed may affect a restoration's bonding and mechanical performance, which are important parameters in understanding clinical behavior.[26] Currently, many options are available to perform glass-ceramic silanization, and manufacturers recommend that they be employed indistinctly (whether silane is mixed with other components or not). To the authors' knowledge, no previous study has evaluated the effect of silane-containing universal adhesives on biaxial flexural strength of a glass-ceramic/resin cement system or the bonding stability they provide to the adhesive interface when submitted to loading forces.

Here we determine whether the use of a silane-containing universal adhesive such as silane primer affects the biaxial flexural strength (BFS) and bonded interface integrity of glass-ceramic/resin cement systems after loading. The null hypotheses tested were that silane-containing universal adhesive does not influence 1) a glass-ceramic/resin cement system's biaxial flexural strength or 2) a glass-ceramic/resin cement system's adhesive interface stability after loading.

METHODS AND MATERIALS

Specimen Fabrication and Group Division

Lithium disilicate CAD/CAM blocks (IPS e.max lithium disilicate CAD/CAM, A2 color, Ivoclar, Vivadent, Schaan, Liechtenstein) were milled on an E4D Dentist System using a cylindrical custom-milled file measuring 6.5 ± 0.1 mm in diameter. Each cylinder was then cut with a diamond saw under water irrigation to obtain discs of 0.5 ± 0.1 mm thickness until completing 120 disc-shaped specimens. Sample measurements were then matched to the appropriate diameter (6.5±0.1 mm) using a digital caliper to fit the biaxial flexure jig. Specimen and flexure jig device dimensions were chosen to simulate the approximate size of a ceramic veneer, as employed in a previous work.[27] Discs were fired unglazed according to the manufacturer's instructions and then ground using #1000 and #2000 grit silicon carbide grinding paper etched with 5% hydrofluoric acid (HF) (Power C Etching, BM4, Palhoça, SC, Brazil) for 20 seconds, water-cleaned for 60 seconds, and ultrasonically cleaned for 5 minutes.

Four groups were formed employing different silanization protocols, treating specimens as follows: 1) RCP (conventional silane): One coat of RelyX Ceramic Primer (3M ESPE, St Paul, MN, USA), a one-bottle conventional silane primer, was actively applied onto the ceramic surface for 60 seconds, followed by thorough drying (20 seconds) using oil-free air until complete solvent evaporation; 2) RCP+SB (conventional silane plus separate adhesive application): one coat of RCP was also applied onto the ceramic surface for 60 seconds and then thoroughly dried (20 seconds) using oil-free air until complete solvent evaporation. Afterward, an adhesive system (Adper Single Bond Plus, 3M ESPE) was applied in one coat for 15 seconds and air-dried for 5 seconds to evaporate the solvent; 3) SBU: Scotchbond Universal (3M ESPE) was applied onto the ceramic surface in one coat for 20 seconds and air-dried for 5 seconds; 4) C: No silane was used and only the previously described HF etching procedure was performed on this group.

After being silanized, all treated surfaces were resin-cement coated (RelyX Ultimate, 3M ESPE). To do so, treated specimens were fixed by the untreated surface on a thick glass plate with the aid of utility wax. One layer of resin cement was placed on the treated surface with the aid of an auto-mixing tip provided by the manufacturer and a micro brush. A polyester strip and a 0.5-mm glass slide were placed on top of the resin cement and pressed using standardized weight devices (200 g) at each side of the glass slide, and at the same time, attached to a digital caliper to control specimen thickness. The resin cement layer was polymerized for 40 seconds (Elipar, S10, 3M ESPE; 800mW/cm^2 as determined using an Ophir Laser measurement potentiometer from Ophir Optronics Ltd., Jerusalem, Israel and taking into account light tip circular area). In the case of groups treated with adhesive systems (RCP+SB and SBU), adhesive and resin cement layers were polymerized simultaneously. Specimen thicknesses were measured again using a digital caliper, and the readings were recorded to calculate each layer's thickness for each specimen (all thicknesses were confirmed using the fractured fragments with the aid of a scanning electron microscope (SEM). Calibration of specimens to fit the flexure jig was performed using a 2000-grit silicon carbide grinding paper whenever necessary. Materials used are described in Table 1.

Biaxial Flexural Strength Test

To measure BFS, the piston-on-ring method was used, employing a customized flexure jig.[27] A schematic representation of the biaxial flexural strength test used in this study is presented in Figure 1. After 24 hour storage in 100% relative humidity at 37°C, all specimens were loosely fitted (resin cement layer facing down) on a support ring (5 mm internal diameter) through a circular aperture (7 mm diameter) of a cylindrical stainless steel jig. Slight specimen flatness imperfections were offset by using a thin piece of rubber film along with a wet piece of filter paper.[28,29] The assembly was positioned on a universal testing machine working at 1.27 mm/min (Instron 4411, Instron Corp, Canton, MA, USA), and a vertical load was applied on the middle of the specimen until failure by a circle-shaped flat piston. The process was monitored and the load recorded at the point of failure, which was used to calculate the BFS/σ_θ according to the bilayer disk approach (considering ceramic and cement as

Table 1: *Materials Used and Their Application Steps*

Material	Type of Material	Manufacturer Lot No.	Composition*	Application Steps
IPS e.max CAD	Lithium disilicate glass-ceramic, A2	Ivoclar, Vivadent, Schaan, Liechtenstein/N76665	SiO_2 , Li_2O, K_2O, P_2O_5, ZrO_2, ZnO, Al_2O_3, MgO, coloring oxides	
RelyX Ceramic Primer (RCP)	Ceramic primer (silane)	3M ESPE St Paul, MN, USA/N406850	MPS, ethanol, water	Apply actively for 60 s, then thoroughly air-dry
Adper Single Bond Plus (SB)	Total-etch adhesive system	3M ESPE Sumaré, SP, Brazil/N334650BR	Bis-GMA, HEMA, dimethacrylates, ethanol,water, photoinitiators, a methacrylate functional copolymer of polyacrylic and polyitaconic acids, and silica nanofiller	Apply actively for 15 s and air-dry for 5 s
Scotchbond Universal (SBU)	Multi-mode adhesive system	3M ESPE St Paul, MN, USA/Neuss, Germany/504115	MDP, dimethacrylate resins, HEMA, Vitrebond TM Copolymer, filler, ethanol, water, initiators, silane	Apply actively for 20 s and air-dry for 5 s
RelyX Ultimate	Composite cement, A2	3M ESPE St Paul, MN, USA/Neuss, Germany/505370	Base paste: Methacrylate monomers, radiopaque silanated fillers, initiator, stabilizer, rheological additives Catalyst paste: Methacrylate monomers, radiopaque alkaline (basic) fillers, initiator, stabilizer, pigments, rheological additives, fluorescent dye, dark cure activator for Scotchbond Universal	Apply the composite cement with an automixing tip (provided by manufacturer) without separating it from the dispensed mass

** Product composition according to materials safety data sheets (MSDS) provided by the manufacturers.*
MPS, methacryloxypropyltrimethoxysilane (prehydrolyzed silane); MDP, 10-methacryloyloxydecyl dihydrogen phosphate; HEMA, 2-hydroxyethyl methacrylate; Bis-GMA, bisphenol A-diglycidyl ether dimethacrylate.

different layers). This method was proposed by Hsueh and others,[29-31] using the analytical model of bilayered disks tested on piston-on-ring device, described by equations 1 to 4[30]:

$$\sigma_\theta = \frac{-PE_2(1+\nu)(z-z_n^*)}{8\pi(1-\nu_2^2)D^*} \times [1 + 2\ln\left(\frac{a}{c}\right) + \frac{1-\nu}{1+\nu}\left(1-\frac{c^2}{2a^2}\right)\frac{a^2}{R^2}] \quad (1)$$

(for $t_1 \leq z \leq t_1 + t_2$ and $r = c$),

where P is the load (N) at fracture, E_2 is the individual Young modulus of layer 2: ceramic (102.7 GPa[32]), z is the axial position of the desired point of calculation on the vertical axis (in this case the axial position used was $z = t_1$ [ceramic/cement interface]), a is the support ring radius (2.5 mm), c is the radius of the indenter of the piston (0.8 mm), and R is the specimen radius (3.25 mm) (Figure 1). The variable ν is given by

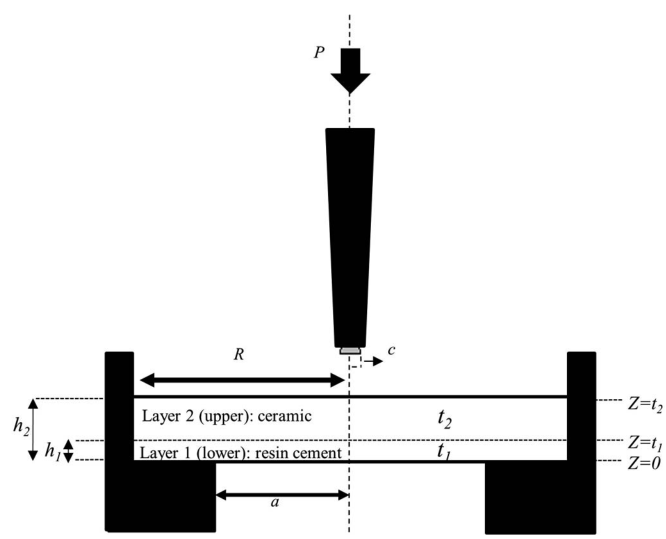

Figure 1. *Schematic representation of the piston-on-ring biaxial flexure test used in this study. Symbology:* P *(load at failure point),* R *(specimen radius),* c *(radius of the indenter of the piston),* t_1 *(individual resin cement layer thickness),* t_2 *(individual ceramic layer thickness),* a *(support ring radius),* z *(axial position).*

Table 2: *Biaxial Flexural Strength Values (BFS) With Standard Deviation (SD) and Weibull Modulus* (m) *With Confidence Intervals (CI) From All Experimental Groups*

Silane Treatment	BFS (SD) (MPa)[a]	*m* (CI)[b]
RCP	364.2 (29.5) A	13.2 (10.1-17.3)
RCP+SB	372.2 (29.4) A	13.1 (10.1-17.0)
SBU	338.0 (27.1) B	16.6 (12.3-22.4)
C	320.7 (36.3) B	9.8 (7.4-12.9)

[a] *Different capital letters represent statistical differences in BFS among the treatments (Tukey,* $p \leq 0.05$*).*
[b] *For* m*, no differences were found. Symbology: Biaxial flexural strength (BFS), standard deviation (SD), confidence interval (CI), RelyX Ceramic Primer (RCP), RelyX Ceramic Primer, and Adper Singlebond Plus (RCP+SB), Scotchbond Universal (SBU), and control (C).*

$$\nu = \frac{\nu_1 t_1 + \nu_2 t_2}{t_1 + t_2} \tag{2}$$

which is an average considering Poisson ratios from each material (0.215 for ceramic [v_2][specifically for E.max CAD[32]], and 0.27 for resin cement [v_1][33]) and each layers' thickness (t_1: individual resin cement layer thickness; t_2: individual ceramic layer thickness). Each specimen's thickness was measured individually. Variable z^* represents the position of the neutral plane and is given by

$$z_n^* = \frac{\frac{E_1 t_1^2}{2(1-\nu_1^2)} + \frac{E_2 t_2^2}{2(1-\nu_2^2)} + \frac{E_2 t_1 t_2}{1-\nu_2^2}}{\frac{E_1 t_1}{1-\nu_1^2} + \frac{E_2 t_2}{1-\nu_2^2}} \tag{3}$$

where E_1 is the Young's modulus of layer 1: resin cement (10 GPa[28]) and variables t_1, t_2, v_1, v_2 and E_2 are the same as used in equations 4 and 5. D^* is the flexural rigidity, described by

$$D^* = \frac{E_1 t_1^3}{3(1-\nu_1^2)} + \frac{E_2 t_2^3}{3(1-\nu_2^2)} + \frac{E_2 t_1 t_2 (t_1 + t_2)}{1-\nu_2^2} - \frac{\left[\frac{E_1 t_1^2}{2(1-\nu_1^2)} + \frac{E_2 t_2^2}{2(1-\nu_2^2)} + \frac{E_2 t_1 t_2}{1-\nu_2^2}\right]^2}{\frac{E_1 t_1}{1-\nu_1^2} + \frac{E_2 t_2}{1-\nu_2^2}} \tag{4}$$

All fragments were collected, identified by specimen, and the number of fragments obtained from each sample was recorded.

Statistical Analysis

Data normality and homoscedasticity were assessed using the Anderson-Darling and Bartlett tests, both at a preset alpha of 0.05. Results were statistically analyzed using a one-way ANOVA (silane treatment) followed by the Tukey pairwise *post hoc* test, performed at a preset alpha of 0.05. Weibull parameters and distribution plots were also generated (Minitab v18.1, Minitab Inc, State College, PA, USA).

Fractured Fragment Interface Analysis

Fractured fragments were mounted on aluminum stubs, sputter coated with gold/palladium (SCD 050; Balzers, Schaan, Liechtenstein), and then examined using a SEM (JSM 5600 LV; JEOL, Tokyo, Japan), operating at 15 kV and a working distance of 20 mm. Images of representative areas of each fragment were obtained to evaluate interfacial characteristics for each group. In addition, each layer's thickness was measured to confirm initial measurements taken during specimen preparation and recorded using the SEM software.

RESULTS

Statistical analysis showed that the data were normally distributed (Anderson-Darling test [p=0.325]); also, homoscedasticity was proved (Bartlett test [p=0.426]), both at a preset alpha of 0.05, indicating allowable use of parametric methods for data analysis. One-way ANOVA statistical analysis revealed that the factor "silane treatment" significantly influenced BFS (p<0.0001). RCP+SB showed the highest BFS mean, showing no statistical difference from the one obtained by RCP (Table 2). SBU presented a lower BFS mean value than RCP+SB and RCP, but not different from C (Table 2).

No statistical differences were found in m, as all confidence intervals overlapped at least at one point (Table 2). Although not statistically significant, the highest m/graph slope was obtained by SBU and the lowest m/graph slope by the C group (Table 2 and Figure 2).

Representative images from fractured specimen analysis are summarized in Figures 3 to 6. Fractured fragment analysis revealed ceramic-cement separation for groups SBU and C (Figures 5 and 6). In the particular case of SBU (Figure 5), the failure line was located mostly between the adhesive layer and the ceramic material. Conversely, groups RCP and RCP+SB showed a uniform interlocking area (resin cement/ceramic [RCP] and resin cement/adhesive/ceramic [RCP+SB/RC]), in which no gap or interruption of continuity were noted (Figures 3 and 4). Additionally, some voids were noticeable within the resin cement layer of all groups.

DISCUSSION

This study shows that silanization using a silane-containing universal adhesive produced lower BFS

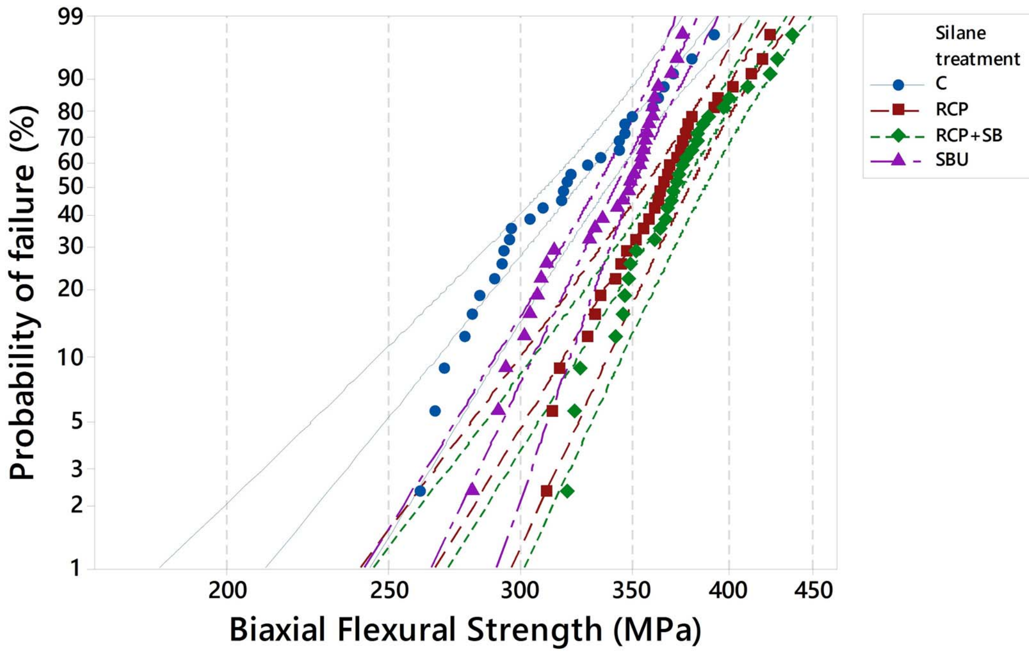

Figure 2. *Weibull distribution corresponding to BFS data. RCP: One-bottle silane primer (RelyX ceramic primer); RCP+SB: Silane and separate application of adhesive (RelyX Ceramic Primer/Adper Single Bond Plus); SBU: Silane-containing universal adhesive (Scotchbond Universal); C: negative control. Lines drawn represent the Weibull curve shape for each group.*

values on ceramic/cement systems than conventional silane and separate application of silane and adhesive. Consequently, the first null hypothesis must be rejected. Because the use of silane-containing universal adhesive as ceramic primer negatively affected the integrity of ceramic/cement adhesive interface during loading, null hypothesis 2 was also rejected. Thus, it can be said that regarding a glass-ceramic/resin cement assembly's mechanical properties, the way resin cement is bonded to the ceramic material may be a very relevant aspect. Additionally, the role of an efficient silane coupling agent appears to be funda-

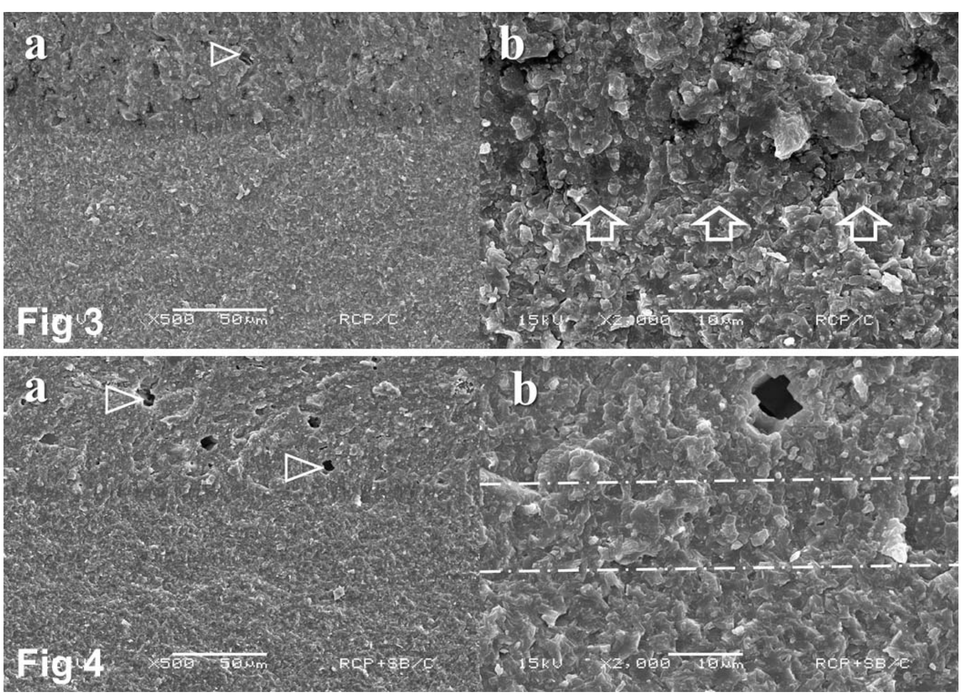

Figure 3. *Representative SEM micrographs of the more prevalent patterns observed in transverse bonded area of the fractured fragment corresponding to group RCP: a) 500 × magnification, showing interlocking of resin cement on ceramic surface and a continuous interface between both materials. Also, some voids within the resin cement layer (triangle pointer), b) close-up from figure 3a (2000×), where ceramic-cement interlocking area is marked between arrows.*

Figure 4. *Representative SEM micrographs of the more prevalent patterns observed in transverse bonded area of the fractured fragment corresponding to group RCP+SB: a) 500× magnification, showing a continuous ceramic-adhesive-cement interlocking and some voids (triangle pointers) within the resin cement layer, b) close-up of Figure 4a (2000 ×), approximate region where ceramic-adhesive and adhesive-cement interlocking areas are located.*

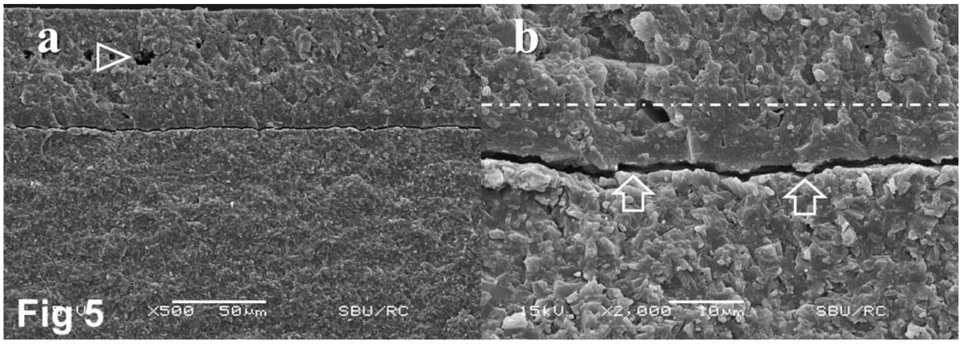

Figure 5. *Representative SEM micrographs of the more prevalent patterns observed in transverse bonded area of the fractured fragment corresponding to group SBU: a) 500 × magnification, showing a separation between the adhesive layer and ceramic material and some voids within resin cement layer (triangle pointer; (500×), b) close-up from Figure 5a (2000×), showing in greater detail the adhesive-cement (pointed line) and ceramic-adhesive interfaces (arrows).*

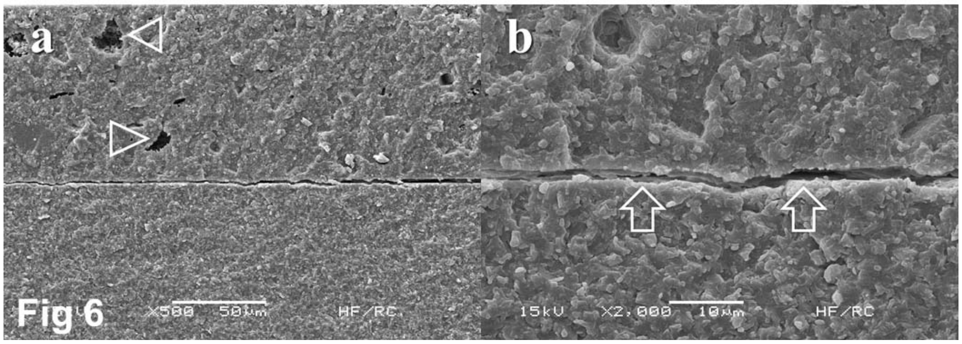

Figure 6. *Representative SEM micrographs of the more prevalent patterns observed in transverse bonded area of the fractured fragment corresponding to group C: a) 500× magnification exhibiting some voids within the resin cement layer (triangle pointers), b) close-up of Figure 6a (2000×), showing a clear separation between the resin cement and the ceramic material (arrows).*

mental in maintaining the integrity of the materials composing the ceramic indirect restoration system.

In order to analyze the mechanical properties of a glass-ceramic/resin cement system influenced by different types of silane primers, the biaxial flexural strength was evaluated using a piston-on-ring biaxial flexure strength test. The biaxial flexure test is more reliable than uniaxial tests, as it applies the stress on a concentric point of the specimen, resulting in a more uniform analysis of material strength.[24,34] The traditional approach for this kind of bending test is not useful to calculate the BFS of multilayered specimens composed of more than one dissimilar material, as it fails to consider the Poisson ratio and individual thickness of each material.[29-31] The analytical solution proposed by Hsueh and others[28-30] includes this possibility, and it is also efficient in calculating biaxial stresses through ceramic/cement bilayered specimens.[28] In the particular case of ceramic specimens treated with an adhesive system layer before resin cement coating (RCP+SB and SBU), although these specimens are composed of three different materials, they were treated as bilayered specimens for calculation purposes, as adhesive layer thickness in this study was recorded to be under 15 μm in all specimens. Such a small thickness would not influence the calculation outcomes.[28] Furthermore, as adhesive and resin cement layers are both composed of resin-based materials and were polymerized together, they were observed to be well integrated; sometimes it was even difficult to differentiate between the two. Thus, being equally valid to apply the bilayer approach for those situations,[24,28] adhesive layers were considered as part of resin cement layers in such cases.

The application of conventional silane (alone or adhesive coated) improved BFS, indicating that silane efficiency in enhancing ceramic/cement bonding plays an important role in a ceramic/cement assembly's mechanical properties. This effect might be explained by the fact that silane bonds chemically with glass-ceramic and resin-based materials, maintaining the integrity of the system. This can be confirmed on SEM images, where a full continuity of the ceramic-cement or ceramic-adhesive-cement interfaces is observed (Figures 3 and 4), suggesting this chemical union was successful. This uniform interlocking area between ceramic and luting materials was previously reported to strengthen glass-ceramics.[21,25] Addison and colleagues suggest that, as an explanation to this phenomenon, when a crack is filled by the resin-based material and the whole system is submitted to load, the Poisson effect is expected to occur, producing a slight contraction at the bottom of the crack (due to its geometry), raising the elastic modulus of the resin-based material in this area and equalizing its behavior to that of the ceramic material.[21,25] This cumulative effect within adjacent cracks may strengthen the resin-based material, maintain ceramic/resin unity, and as a consequence, increase the flexural strength of the whole system.[21,25] On the other hand, applying an adhesive coat after the silane primer may present better wetting and interpenetrating capabilities than would the resin cement due to the adhesive's lower filler content and viscosity, resulting in a better intimacy with the ceramic material.[23,35] Thus, in light of our results, adhesive coating of previously silanized glass-ceramics has a positive effect on the ceramic/cement system's mechanical properties.

Silane-containing universal adhesives are also recommended by manufacturers to be used as silane primers. In the present study, this kind of material demonstrated no positive effect on the biaxial flexural strength, as it showed no statistical difference compared with the negative control group (Table 2). This may be due to some kind of inefficient bonding property demonstrated by the silane contained in these adhesives, incapable of maintaining the integrity of the ceramic-adhesive-cement assembly, as shown in Figure 5. This lack of unity between "bonded" materials may lead to a faster propagation of microcracks, consequently weakening the whole specimen.

In this study, this pattern was observed in the SBU group and in the negative control group in which silane was not applied (Figures 5 and 6). Previous studies have demonstrated that conventional silane (with or without separate applications of adhesive) performs better than silane-containing universal adhesives as a ceramic-cement bonding promoter.[17-19] Universal adhesives contain many ingredients other than silane, resulting in fewer silane molecules per area in contact with the ceramic surface,[36] in contrast to the silane-only containing primer. Intimate contact between the silane and ceramic surface is crucial, as one silane coat contains three oligomer layers,[37] just the first being capable of forming chemical bonds; the outermost layers may be detrimental.[38] Also, elimination of solvents and other byproducts formed during the silane condensation reaction may be hindered through development of a dense polymer network,[39] needing more time to evaporate solvent in universal adhesives as demonstrated in a previous work.[40] Moreover, a more acidic environment of universal adhesives (SBU, pH 2.7; RCP, pH 4.6)[2] may lead to continuous hydrolyzation and reaction of its silane molecules upon storage, and consequently being inactive to some degree before being used, as proven by Yoshihara and others.[20]

Additionally, the type of silane contained in SBU is not specified; it is possible that the silane compound used in those adhesives is not as effective as the well-known methacryloxypropyltrimethoxysilane. All these issues may explain why universal adhesives failed to improve the ceramic/cement system's BFS and maintain its integrity with resin cement coating. However (though not statistically significant), the SBU attained the highest m (16.6) among all groups (Figure 2, Table 2), being very distant from the C group in this regard. This may be explained by the fact that, despite the similar behavior that they (SBU and C) showed regarding BFS and adhesive interface integrity (Table 2, Figures 5 and 6), SBU as an adhesive may have better wettability than the resin cement alone and, further, some positive effect (even though low) would be expected from SBU compared with the control group.

Previous studies have stated that resin cement coatings increase the BFS of ceramic materials,[35,41] while silane priming itself does not.[25] Based on our results, it cannot be said that silane enhances ceramic BFS, but silane's effectiveness does affect adhesive interface behavior of a ceramic/cement assembly during load and consequently its mechanical properties. This scenario can be extrapolated to a clinical situation in which an indirect all-ceramic restoration is luted with resin cement without treating the internal ceramic surface using an efficient silane primer. In that case, less likelihood of restoration success can be expected, as shown here by m (Figure 2, Table 2). Thus, we may infer that a positive effect of a resin-cement coating on a ceramic/cement assembly's BFS depends on the performance of a proper silanization process. Resin cement coating per se is no guarantee of improved mechanical properties on ceramic/cement systems, as some voids were found across the resin cement layer in all groups (Figures 3 to 6), even though an auto-mixing tip was used, avoiding manual manipulation. Thus, a "perfect" resin cement layer (with no such defects) may be difficult to reproduce in clinical situations, comprising the mechanical reliability of the restoration.

As shown in this study, glass-ceramic/resin cement chemical bonding seems to be crucial not just for bond strength but for a ceramic cement system's mechanical properties, as the integrity of a bilayered system may be considered a more important factor than the strengthening potential of each layer by itself.

CONCLUSIONS

Within the limitations of this *in vitro* study, the following conclusions can be drawn:

1. The silanization protocol influences the biaxial flexural strength of glass-ceramic/resin cement systems.

2. The application of a silane primer (alone or adhesive coated) to the ceramic surface improves the biaxial flexural strength of glass-ceramic/resin cement assemblies, while the application of a silane-containing universal adhesive does not.

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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 presented in this article.

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REFERENCES

1. Peumans M, Van Meerbeek B, Lambrechts P, & Vanherle G (2000) Porcelain veneers: A review of literature *Journal of Dentistry* **28(3)** 163-177.
2. Ying Kei Lung C, & Matinlinna JP (2012) Aspects of silane coupling agents and surface conditioning in dentistry: An overview *Dental Materials* **28(5)** 467-477.
3. Peumans M, De Munck J, Van Landuyt KL, Poitevin A, Lambrechts P, & Van Meerbeek B (2010) Eight-year clinical evaluation of a 2-step self-etch adhesive with and without selective enamel etching *Dental Materials* **26(12)** 1176-1184.
4. Brentel SA, Özcan M, Valandro LF, Guimarães Alar?a L, Amaral R, & Bottino MA (2007) Microtensile bond strength of a resin cement to feldspathic ceramic after different etching and silanization regimens in dry and aged conditions *Dental Materials* **23(11)** 1323-1331.
5. Blatz MB, Dent M, Sadan A, & Kern M (2003) Resin-ceramic bonding: A review of the literature *Journal of Prosthetic Dentistry* **89(3)** 268-274.
6. Scherrer SS, de Rijk WG, Belser UC, & Meyer JM (1994) Effect of cement film thickness on the fracture resistance of a machinable glass-ceramic *Dental Materials* **10(3)** 172-177.
7. Fradeani M, & Redemagni M (2002) An 11-year clinical evaluation of leucite-reinforced glass-ceramic crowns: A retrospective study *Quintessence International* **33(7)** 503-510.
8. Chen JH, Matsumura H, & Atsuta M (1998) Effect of etchant, etching period, and silane priming on bond strength to porcelain of composite resin *Operative Dentistry* **23(5)** 250-257.
9. Addison O, Marquis PM, & Fleming GJ (2007) The impact of hydrofluoric acid surface treatments on the performance of a porcelain laminate restorative material *Dental Materials* **23(4)** 461-468.
10. Matinlinna JP, Lassila LV, & Vallittu PK (2007) Pilot evaluation of resin composite cement adhesion to zirconia using a novel silane system *Acta Odontologica Scandinavica* **65(1)** 44-51.
11. Matinlinna JP, Lassila LV, Özcan M, Yli-Urpo A, & Vallittu PK (2004) An introduction to silanes and their clinical applications in dentistry *International Journal of Prosthodontics* **17(2)** 155-164.
12. Anagnostopoulos T, Eliades G, & Palaghias G (1993) Composition, reactivity and surface interaction of three dental silane primers *Dental Materials* **9(3)** 182-190.
13. Blatz MB, Sadan A, Martin J, & Lang B (2004) In vitro evaluation of shear bond strengths of resin to densely sintered high-purity zirconium-oxide ceramic after long-term storage and thermal cycling *Journal of Prosthetic Dentistry* **91(4)** 356-362.
14. Kitayama S, Nikaido T, Takahashi R, Zhu L, Ikeda M, Foxton RM, Sadr A, & Tagami J (2010) Effect of primer treatment on bonding of resin cements to zirconia ceramic *Dental Materials* **26(5)** 426-432.
15. Van Meerbeek B, Yoshihara K, Yoshida Y, Mine A, De Munck J, & Van Landuyt KL (2011) State of the art of self-etch adhesives *Dental Materials* **27(1)** 17-28.
16. Van Meerbeek B, De Munck J, Yoshida Y, Inoue S, Vargas M, Vijay P, Van Landuyt K, Lambrechts P, & Vanherle G (2003) Buonocore memorial lectura. Adhesion to enamel and dentin: Current status and future challenges *Operative Dentistry* **28(3)** 215-235.
17. Kalavacharla VK, Lawson NC, Ramp LC, & Burgess JO (2015) Influence of etching protocol and silane treatment with a universal adhesive on lithium disilicate bond strength *Operative Dentistry* **40(4)** 372-378.
18. Kim RJ, Woo JS, Lee IB, Yi YA, Hwang JY, & Seo DG (2015) Performance of universal adhesives on bonding to leucite-reinforced ceramic *Biomaterials Research* **22(19:11)** 1-6.
19. Murillo-Gómez F, Rueggeberg FA, & De Goes MF (2017) Short- and long-term bond strength between resin cement and glass-ceramic using a silane-containing universal adhesive *Operative Dentistry* **42(5)** 514-525.
20. Yoshihara K, Nagaoka N, Sonoda A, Maruo Y, Makita Y, Okihara T, Irie M, Yoshida Y, & Van Meerbeek B (2016) Effectiveness and stability of silane coupling agent incorporated in 'universal' adhesives *Dental Materials* **32(10)** 1218-1225.
21. Addison O, Marquis PM, & Fleming GJ (2007) Resin elasticity and the strengthening of all-ceramic restorations *Journal of Dental Research* **86(6)** 519-523.
22. Addison O, Marquis PM, & Fleming GJ (2008) Quantifying the strength of a resin-coated dental ceramic *Journal of Dental Research* **87(6)** 542-547.
23. Fleming GJ, Hooi P, & Addison O (2012) The influence of resin flexural modulus on the magnitude of ceramic strengthening *Dental Materials* **28(7)** 769-776.
24. Posritong S, Borges AL, Chu TM, Eckert GJ, Bottino MA, & Bottino MC (2013) The impact of hydrofluoric acid etching followed by unfilled resin on the biaxial strength of a glass-ceramic *Dental Materials* **29(11)** e281-e290.
25. Addison O, Marquis PM, & Fleming GJ (2007) Resin strengthening of dental ceramics—the impact of surface texture and silane *Journal of Dentistry* **35(5)** 416-424.

26. Albakry M, Guazzato M, & Swain MV (2004) Biaxial flexural strength and microstructure changes of two recycled pressable glass ceramics *Journal of Prosthodontics* **13(3)** 141-149.

27. Rueggeberg FA, Cole MA, Looney SW, Vickers A, & Swift EJ (2009) Comparison of manufacturer-recommended exposure durations with those determined using biaxial flexure strength and scraped composite thickness among a variety of light-curing units *Journal of Esthetic and Restorative Dentistry* **21(1)** 43-61.

28. Addison O, & Fleming GJP (2008) Application of analytical stress solutions to bi-axially loaded dental ceramic-dental cement bilayers *Dental Materials* **24(10)** 1336-1342.

29. Hsueh CH, Luttrell CR, & Becher PF (2006) Modelling of bonded multilayered disks subjected to biaxial flexure tests *International Journal of Solids and Structures* **43** 6014-6025.

30. Hsueh CH, Luttrell CR, & Becher PF (2006) Analyses of multilayered dental ceramics subjected to biaxial flexure tests *Dental Materials* **22(5)** 460-469.

31. Huang CW, & Hsueh CH (2011) Piston-on-three-ball versus piston-on-ring in evaluating the biaxial strength of dental ceramics *Dental Materials* **27(6)** e117-e123.

32. Wendler M, Belli R, Petschelt A, Mevec D, Harrer W, Lube T, Danzer R, & Lohbauer U (2017) Chairside CAD/CAM materials. Part 2: Flexural strength testing *Dental Materials* **33(1)** 99-109.

33. De Jager N, Pallav P, & Feilzer AJ (2004) The apparent increase of the Young's modulus in thin cement layers *Dental Materials* **20(5)** 457-462.

34. Al-Makramani BM, Razak AA, & Abu-Hassan MI (2010) Biaxial flexural strength of Turkom-Cera core compared to two other all-ceramic systems *Journal of Applied Oral Sciences* **18(6)** 607-612.

35. Spazzin AO, Guarda GB, Oliveira-Ogliari A, Leal FB, Correr-Sobrinho L, & Moraes RR (2016) Strengthening of porcelain provided by resin cements and flowable composites *Operative Dentistry* **41(2)** 179-188.

36. Ikemura K, Tanaka H, Fujii T, Deguchi M, Negoro N, Endo T, & Kadoma Y (2011) Design of a new, multi-purpose, light-curing adhesive comprising a silane coupling agent, acidic adhesive monomers and dithiooctanoate monomers for bonding to varied metal and dental ceramic materials *Dental Materials Journal* **30(4)** 493-500.

37. Ishida H, & Koenig JL (1980) A Fourier-transform infrared spectroscopic study of the hydrolytic stability of silane coupling agents on E-glass fibers *Journal of Polymer Science Part B: Polymer Physics* **18** 1931-1943.

38. Berg J, & Jones FR (1998) The role of sizing resins, coupling agents and their blends on the formation of the interphase in glass fiber composites *Composites Part A: Applied Science and Manufacturing* **29(A)** 1261-1272.

39. Shen C, Oh WS, & Williams JR (2004) Effect of post-silanization drying on the bond strength of composite to ceramic *Journal of Prosthetic Dentistry* **91(5)** 453-458.

40. Luque-Martinez IV, Perdigão J, Muñoz MA, Sezinando A, Reis A, & Loguercio AD (2014) Effects of solvent evaporation time on immediate adhesive properties of universal adhesives to dentin *Dental Materials* **30(10)** 1126-1135.

41. Salazar-Marocho SM, de Melo RM, Macedo LG, Valandro LF, & Bottino MA (2011) Strength of a feldspar ceramic according to the thickness and polymerization mode of the resin cement coating *Dental Materials Journal* **30(3)** 323-329.