BJOS
Brazilian Journal of Oral Sciences

Original Article

Volume 20
2021
e211670

Surface treatment and adhesion approaches on polymer-infiltrated ceramic network: influence on the bond strength to resin cement

Michele Mirian May[1], Camila da Silva Rodrigues[1], Juliane Bortolotto da Rosa[2], Júlia Persio Herrmann[3], Liliana Gressler May[1]*

[1] Post-graduate Program in Dental Sciences, Federal University of Santa Maria.

[2] Residency Program in Professional Health Area, Multiprofessional modality, Federal University of Santa Maria.

[3] Private Dentistry Practice.

Corresponding author:
Liliana Gressler May, DMD, MSD, PhD, Associate Professor, Federal University of Santa Maria, Faculty of Odontology, MSciD-PhD Graduate Program in Dental Science, Prosthodontics Unit.
Avenida Roraima, 1000, prédio 26F, sala 2386, Campus UFSM, Santa Maria, RS, Brasil,
CEP: 97105-900,
Phone number: +55 55 3220 9276 / Fax number: +55 55 3220 9272
e-mail: liligmay@gmail.com

Received: October 16, 2020
Accepted: December 17, 2021

Aim: To evaluate the effect of different surface treatments and adhesive approaches on the microshear bond strength of resin cement to a polymer-infiltrated ceramic network (PICN). **Methods:** PICN blocks were randomly assigned into 9 groups (n=10): CTRL: no treatment; HF: 5% hydrofluoric acid etching; HF-S: HF + silane; HF-S-A: HF-S + adhesive (Adper Single Bond 2); HF-UA: HF + universal adhesive (Single Bond Universal); SB: sandblasting with 50 µm Al_2O_3 particles; SB-S: SB + silane; SB-S-A: SB-S + adhesive; SB-UA: SB + universal adhesive. Resin cement microcylinders (Ø = 0.96 mm; height = 1 mm) (RelyX Ultimate) were built upon the PICN surface after roughness and contact angle measurements. Next, microshear bonding tests (µSBS) were performed (0.5 mm/min) after water storage (37ºC, 90 days) and thermocycling (12,000 cycles; 5ºC-55ºC). Failure modes were observed under stereomicroscope. Bond strength data were analyzed by two-way ANOVA/Tukey's test and t-tests. Kruskal-Wallis/Dunn's tests were conducted for roughness and contact angle data (α = 0.05). **Results:** A rougher surface and lower contact angles were observed for Sandblasting. HF-S (18.54 ± 2.03 MPa), SB-S (19.00 ± 1.66 MPa) and SB-UA (18.07 ± 2.36 MPa) provided the highest bond strength values, followed by the other treated groups. The CTRL group resulted in lower bond strength (7.18 ± 2.34 MPa). **Conclusion:** Hydrofluoric acid etching followed by silane application and sandblasting followed by silane or universal adhesive are useful clinical steps to enhance bonding to PICN. Adhesive applications after HF etching have no advantages in bonding to PICN.

Keywords: Ceramics. Hydrofluoric Acid. Adhesives. Air abrasion, dental. Resin cements. Surface properties.

Introduction

Polymer-infiltrated ceramic network (PICN) is a restorative material for use in CAD/CAM (computer-aided design and computer-aided manufacturing). It promises to combine the qualities of ceramics such as durability, color stability, and the improved flexural properties and low abrasiveness from the resin composites[1-5]. Also known as a hybrid ceramic, Vita Enamic (Vita Zahnfabrik, Bad Säckingen, Germany) consists of a dominant porous feldspathic ceramic matrix (86 wt%) infiltrated with a copolymer (urethane dimethacrylate and triethylene glycol dimethacrylate) (14 wt%) uniformly incorporated with each other[1,2,6-8].

Long-term success of restorations depends on establishing a reliable bond between the restorative material and the luting agent. Furthermore, adhesive bonding is related to a higher fracture strength of indirect restorations and restored teeth[4,9,10]. Methods which increase the surface properties would be clinically advantageous in order to improve the adhesive bond to ceramic surfaces[7,11-13], promoting micromechanical interlocking[14-16] and/or surface reactivity for chemical bonding[17-19].

Bonding between resin cements and PICN are challenging due to the high degree of polymer conversion (up to 96%), with only a few free monomers remaining available for copolymerization with resin cement and its specific microstructure[3,13,15,16]. Hydrofluoric acid etching and sandblasting with alumina particles/silica coating followed by silane application have been evaluated for PICN[10,12,13,20-23]. However, literature is controversial about the aforementioned approaches. While some authors report improved bond strength when HF acid etching was used[24], others suggest better results from sandblasting treatments[12,25], or even no difference when using any of these alternatives[11,26].

Silane coupling agent has demonstrated a positive impact on the bond strength of composite cements to PICN[4,19,26,27]. However, only the ceramic component of PICN has a chemical bond to silane, as the resinous component presents limited reactive groups available for bonding after polymerization[22]. A universal adhesive containing methacrylate-modified polyalkenoic acid copolymers, methacryloyloxydecyl dihydrogen phosphate (MDP) and silane might be capable to bind to both phases (ceramic and resin) of the polymer-infiltrated ceramic network[22]. In addition, this universal adhesive can react with the dental structure[28]. Thus, the use of these multi-mode systems enables a wide range of applications combined in a single product, therefore being an economical alternative for dentists. Although the latest instructions brochure of PICN[6] recommends the use of silane, the International Academy for Adhesive Dentistry suggested that the adhesive application after silanization could improve resin infiltration within the etched surface[7]. Furthermore, a systematic review and meta-analysis found that the highest bond strength values for PICN ceramic materials is provided by chemical etching followed by a universal primer application[13].

Given the above, the present work aims to evaluate the effect of different surface treatments of PICN for mechanical interlocking (hydrofluoric acid etching vs alumina sandblasting) and adhesive procedures - silane, silane followed by adhesive (2 steps),

MDP containing adhesive with silane (1 step, single bottle, "universal adhesive") - on the bond strength to resin cement after aging. The hypotheses were: 1) PICN surface treatments followed or not by adhesive application (use of coupling agents) would increase the bond strength to resin cement compared to no treatment; 2) hydrofluoric acid etching and alumina sandblasting would behave similarly with respect to resin cement bonding; 3) the use of coupling agents for chemical conditioning following PICN surface treatments would increase its bond strength to resin cement; and 4) the different coupling agents following surface treatments would have similar effects on the bond strength results.

Materials and methods

Table 1. Composition of the used materials

Material	Composition	Manufacturer	Batch number
VITA ENAMIC; polymer- infiltrated ceramic network material	Feldspathic ceramic (86 wt%), polymer (14 wt%) Hybrid ceramic (resin infiltrated ceramic network) Ceramic: silicon dioxide 58–63%, aluminum oxide 20–23%, sodium oxide 9–11%, potassium oxide 4–6%, boron trioxide 0.5–2%, zirconia and calcium oxide. Polymer part (25%): UDMA and TEGDMA	VITA Zahnfabrik, Bad Säckingen, Germany	40370
Al_2O_3 powder	50 µm aluminum oxide	Bio-Art Equipamentos Odontológicos Ltda, São Carlos, SP, Brazil.	52160
Condac Porcelana	5% hydroofluric acid gel	Dentscare Ltda. (FGM), Joinville, SC, Brazil.	161117
RelyX Ceramic Primer	Ethyl alcohol, water and methacryloxypropyltrimethoxysilane	3M ESPE, St. Paul, USA	N878550
Adper Single Bond 2	BisGMA, HEMA, UDMA, dimethacrylates, ethanol, water, glycerol, photoinitiators methacrylate copolymer of polyacrylic and polyitaconic acids, silica nanofiller treated with silane	3M ESPE, Sumaré, SP, Brazil	N895742
Single Bond Universal Adhesive	BisGMA, HEMA, decamethylene dimethacrylate, ethanol, water, silane-treated silica, 2-propenoic acid, methacrylated phosphoric acid, copolymer of acrylic and itaconic acid, ethyl-4-dimethylaminobenzoat, camphorquinone, (dimethylamino) ethyl methacrylate, methyl ethyl ketone, MDP, silane	3M ESPE, Sumaré, SP, Brazil	663003
RelyX Ultimate; dual- polymerizing resin cement	MDP, silanated fillers, ethanol, Vitrebond copolymer, HEMA, initiator components, dimethacrylate resins, water Base paste: Silane–treated glass powder, 2-propenoic acid, 2-methyl-, reaction products with 2-hydroxy-1,3-propanedyl dimethacrylate and phosphorus oxide, TEGDMA, silane-treated silica, oxide glass chemicals, sodium persulfate, tertbutyl peroxy-3,5,5-trimethylhexanoate, copper acetate monohydrate; Catalyst paste: Silane-treated glass powder, substituted dimethacrylate, 1,12-dodecane dimethacrylate, silane--treated silica, 1-benzyl-5-phentyl-barbic-acid, calcium salt, sodium p-toluenesulfinate, 2-propenic acid, 2-methyl-, di-2,1-ethanediyl ester, calcium hydroxide, titanium dioxide	3M ESPE, Seefeld, Germany	1726200736

Table 2. Experimental Design

Mechanical conditioning	Chemical conditioning	Group (n=10)
No surface treatment	-	CTRL
5% Hydrofluoric acid etching	-	HF
	Silane	HF-S*
	Silane + Single Bond 2	HF-S-A
	Single Bond Universal	HF-UA
50 µm alumina sandblasting	-	SB
	Silane	SB-S
	Silane + Single Bond 2	SB-S-A
	Single Bond Universal	SB-UA

*Surface treatment recommended by the manufacturer

The factors evaluated in this in vitro study were surface treatment (etching with 5% hydrofluoric acid, sandblasting or no treatment – control) and coupling agent (only silane, silane + adhesive, or only universal adhesive). The main outcome variable analyzed was microshear bond strength. The materials used in this study, their compositions, commercial names and manufacturers are described in Table 1. The experimental design is shown in Table 2.

Five blocks of a PICN (Vita Enamic, Vita Zahnfabrik, Bad Säckingen, Germany) were cut into 90 slices (6x7x1.5 mm^3) with a diamond saw in a cutting machine (Labcut 1010, Extec Co, Enfield, USA). The plates were polished with 600 grit silicon carbide paper under water cooling to standardize the surfaces. The specimens were subsequently placed in individual packages, numerically sorted and randomly assigned into 9 groups conducted by a random sequence generation program (random.org).

The ceramic slices were centrally positioned in plastic cylinders (14 mm high and 25 mm in diameter) and embedded into self-curing acrylic resin. The cementation surface was kept free during embedding. All the specimens were ultrasonically cleaned (1440 D–Odontobras, Ribeirao Preto, SP, Brazil) in distilled water for 5 minutes, dried and cleaned with 99.3% ethanol (Rioquímica, São José do Rio Preto, SP, Brazil) using a disposable microtip applicator followed by air-drying for 20 seconds. The cementation surfaces of the specimens were then treated according to the following approaches (n=10): Control (**CTRL**): The ceramic blocks received no surface treatment (negative control); Hydrofluoric acid etching (**HF**): 5% hydrofluoric acid gel (Condac Porcelana, FGM, Joinville, SC, Brazil) was applied to the adhesive surface for 60 s. Then the acid residues were removed with water spray, followed by an ultrasonic bath in distilled water for 5 min to remove debris and precipitates, cleaning with 99.3% alcohol[7] using a disposable microtip applicator, and air-drying for 20 seconds. This protocol follows the manufacturer's recommendation[6]; Sandblasting (**SB**): The adhesive surface was sandblasted with aluminum oxide particles (50 µm) (Bioart, São Carlos, SP, Brazil) at a distance of 10 mm perpendicular to specimen and a pressure of 2 bar for 10 seconds. A device was used to standardize the application. After sandblasting, the specimens were ultrasonically cleaned

in distilled water for 5 minutes, dried and cleaned with 99.3% ethanol using a disposable microtip applicator and air-dried for 20 seconds; Silane: **HF-S** and **SB-S** received a silane coupling agent application (RelyX Ceramic Primer, 3M ESPE, St. Paul, USA) after their respective surface treatment. The silane was then scrubbed for 60 seconds using a disposable microtip applicator, gently air-dried for 5 seconds and allowed to react for 5 minutes; Silane + adhesive: The same protocol as the HF-S and SB-S was conducted for **HF-S-A** and **SB-S-A**, respectively. Next, the Adper Single Bond 2 adhesive (3M ESPE, Sumaré, SP, Brazil) was applied onto the surface for 20 seconds with a disposable microtip applicator and gently air-dried for 5 seconds; Universal adhesive: After the respective surface treatments for the **HF-UA** and **SB-UA** groups, the Single Bond Universal adhesive (3M ESPE, Sumaré, SP, Brazil) was actively applied onto the surface with a disposable microtip applicator for 20 seconds and gently air-dried for 5 seconds.

The roughness of the top surface of the specimens was measured at the end of each surface treatment (no treatment, hydrofluoric etching and sandblasting) using a contact stylus profilometer (SJ-410, Mitutoyo, Japan).

The Ra (average surface roughness; μm) and Rz (arithmetic mean peak-to-valley height; μm) values were determined using the average of six measurements (three along a "x" direction and three in a "y" direction), with a cut-off (n=5), λC 0.8 mm and λS 2.5 μm, considering the ISO:4287-1997[29].

A goniometer (Drop Shape analysis, model DSA 30S, Kruss GmbH, Hamburg, Germany) connected to a computer containing a dedicated software (DSA3, V1 .0.3-08, Kruss) to determine the contact angles was used for the measurement. After the surface treatments (no treatment, hydrofluoric etching and sandblasting), the contact angle of all specimens was assessed via the sessile drop technique at room temperature (±24°C). Next, one drop (11 μl) of distilled water was deposited at the center of the hybrid ceramic surface using a needle and the contact angle was measured after 5 seconds[30].

Four starch tubes (1 mm height; 0.96 mm internal diameter; Renata, Pastificio Selmi, Londrina, PR, Brazil) were placed over the previously treated surface of each specimen[31] and fixed with wax. Next, the resin cement (Relyx Ultimate, 3M ESPE, Seefeld, Germany) was applied inside the tubes by the same operator (M.M.) at room temperature (22-24°C) and light-polymerized (Radii-cal, SDI, Bayswater, WA, Australia) for 40 s. The samples were stored in distilled water at 37°C for 24 h. After this period, the starch tubes were carefully removed and the specimens were analyzed using a stereomicroscope (Stereomicroscope Discovery V20, Carl Zeiss, Göttingen, Germany) at 35x magnification to observe the adhesive interface. The microcylinders were discarded if gaps, air bubbles, or other defects were detected.

All the specimens were stored for 90 days before testing in distilled water at 37°C and thermocycled (12,000 cycles; 5-55°C; 30 s dwelling time; 2 s transfer time) (Nova Etica, Vargem Grande do Sul, SP, Brazil).

For microshear testing (μSBS), the samples were mounted on a specific device and connected to a universal testing machine (EMIC DL1000, Emic, São José dos Pinhais, PR, Brazil). A stainless-steel wire (0.3 mm diameter) was placed as close as

possible to the free surface of the PICN, being in contact with the lower half-circle of the resin cement microcylinder. Next, a shear load (load cell 0.1 KN) was applied at a rate of 0.5 mm/min until failure occurred. The bond strength *R* (MPa) was calculated using the equation R = F/A, in which "F" is the load for failure of the microcylinder (N) and "A" is the area of its adhesive interface (mm^2). The adhesive area A (mm^2) was calculated from the cross-sectional area of the cylinder, accessed by $A = pr^2$, in which "p" = 3.1416 and "r" is the radius of the obtained circle (0.48 mm), and was found to be 0.72 mm^2.

Each PICN embedded slice was considered as a sample unit (n=10). The bond strength per specimen was calculated as the average from the bond strength values of their microcylinders after excluding pre-test failures.

The failure mode was determined under a stereomicroscope (Discovery V20, Carl-Zeiss, Berlin, Germany), and classified into 4 types: ADHES) adhesive failure (no cement residues present on the PICN surface); Pred-ADHES) predominantly adhesive (more than 50% of adhesive failure, but there were remnants of cement on the cementation surface); COHES-cem) cohesive failure at the cement; COHES-cer) cohesive failure at the hybrid ceramic. Additionally, representative specimens for failure modes were selected and analyzed by SEM (Vega3, Tescan, Brn, Czech Republic).

The PICN plate was considered as the experimental unit for bond strength data analysis. Thus, the bond strength of each plate was calculated by the average of the values obtained from each resin cement cylinder tested from the plate. The sample size was maintained (n = 10), except for the groups HF-SA (n = 7) and HF-UA (n = 9) due to pre-test failures during thermocycling.

The statistical analysis was performed using the SigmaPlot 12.0 software program (Systat Software Inc, San Jose, CA, USA). Data was previously subjected to homoscedasticity (Levene test) and normality (Shapiro-Wilk test) tests. Bond strength data were first analyzed with Two-way ANOVA (surface treatment*coupling agent) and Tukey's test *post-hoc*. In addition, each experimental group were separately compared to the control group using t-tests. Roughness (Ra and Rz) and contact angle data were analyzed by the Kruskal-Wallis test followed by Dunn's test for multiple comparisons.

Results

Table 3. Means (standard deviations) of average roughness (Ra), ten-point-mean roughness (Rz), and contact angle of the ceramic after the surface treatments.

	Ra (µm)	Rz (µm)	Contact angle (°)
No treatment	0.27 (0.03)^{c}	2.17 (0.26)^{c}	84.07 (11.01)^{a}
Hydrofluoric acid	0.58 (0.04)^{b}	4.32 (0.30)^{b}	73.65 (10.57)^{a}
Sandblasting	1.90 (0.17)^{a}	11.16 (0.75)^{a}	59.29 (5.38)^{b}

Different letters within a column indicate statistical differences between the surface treatments (Kruskall-Wallis and Dunn's tests ($P < 0.05$)).

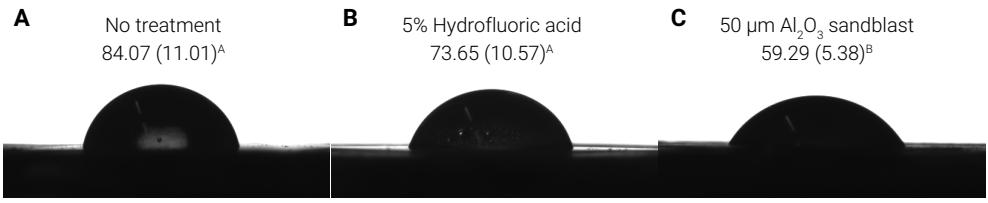

Figure 1. Images and means ± SD (in degrees) of contact angle measurements of surfaces subjected to the following conditions: no treatment (A); 5% hydrofluoric acid etching for 60 s (B); sandblasting with 50 µm alumina particles (C). Different superscript letters indicate statistically significant differences (Kruskall-Wallis and Dunn's tests ($P < 0.05$)

Table 4. Means (standard deviations) of bond strength data (MPa), percentage of pre-tested failures during thermo-cycling, and failure modes of each tested group.

Surface treatment	Bonding agent	Group	Bond Strength (MPa)	Pre-test failure during aging (%)	Failure Modes (%)			
					Adhesive	Predominantly adhesive	Cohesive in ceramic	Cohesive in cement
No surface treatment	None	CTRL	7.19 (2.22)	20.0	64	33	0	3
Hydrofluoric acid	None	HF	13.47 (1.49)^{b*}	0	95	5	0	0
	Silane	HF-S	18.54 (1.93)^{a*}	0	12.5	30	57.5	0
	Silane + Adhesive	HF-S-A	15.40 (2.86)^{b*}	67.5	84.6	7.7	0	7.7
	Universal Adhesive	HF-UA	13.07 (2.05)^{b*}	45.0	100	0	0	0
Sandblasting	None	SB	15.29 (2.39)^{cd*}	0	92.5	5	2.5	0
	Silane	SB-S	19.00 (1.57)^{ab*}	5.0	45	7.5	42.5	0
	Silane + Adhesive	SB-S-A	13.79 (2.53)^{d*}	32.5	100	0	0	0
	Universal Adhesive	SB-UA	18.06 (2.24)^{bc*†}	7.5	89.2	10.8	0	0

Different letters within a column indicate statistical differences between the experimental groups, separately for surfaces treated with hydrofluoric acid or sanblasting (Two-way ANOVA, Tukey's test, $P < 0.05$).
†Difference between HF and S using the same bonding agent.
*Groups that were statistically different from the CTRL (t-test, $P < 0.05$)

The Kruskal-Wallis test indicated statistically significant differences between the roughness values from the different surface treatments ($P < 0.05$). The mean values of roughness parameters and contact angle of the ceramic after surface treatment are summarized in Table 3. Sandblasted PICN showed the highest Ra and Rz values, followed by the etched samples. The CTRL group exhibited the lowest surface roughness.

The etched PICN surface (HF) presented a statistically similar contact angle to the non-treated surface (CTRL). The lowest contact angle values were presented by sandblasted surfaces (SB) ($P < 0.05$) (Table 3). Representative images of the contact angles for the surface treatments for mechanical interlocking can be observed in Fig. 1. The mean µSBS values, percentages of pre-test failures (specimens which failed during the aging process) and failure modes of tested group are compiled in Table 4.

All the treatments resulted in higher bond strength than the CTRL (non-treated surface) (7.19 (±2.22) MPa) (t-test). The highest bond strength values were achieved when only silane coupling agent was applied before cementation for both mechanical treatments (HF-S 18.54 (±1.93) MPa and SB-S 19.00 (±1.57) MPa). All the adhesive procedures were statistically similar when comparing sandblasted and etched groups, except for the universal adhesive groups in which sandblasting followed by universal adhesive presented better performance than HF etching followed by UA (SB-UA 18.06 (±2.24) MPa; HF-UA13.07 (±2.05) MPa), comparable to the application of silane after sandblasting (SB-S) ($P < 0.05$).

The groups with adhesive application (HF-S-A and SB-S-A) as well as HF etching followed by universal adhesive treatment (HF-UA) showed a high percentage of pre-test failures during thermocycling. Pre-test failures in these groups were greater than in the untreated group (CTRL). HF, HF-S and SB groups did not present pre-test failures (Table 4). The microcylinders which exhibited pre-test failure were excluded from the statistical analyses. Adhesive failures were predominant in all groups (except for HF-S), with a majority of cohesive failures in the ceramic (Fig. 2).

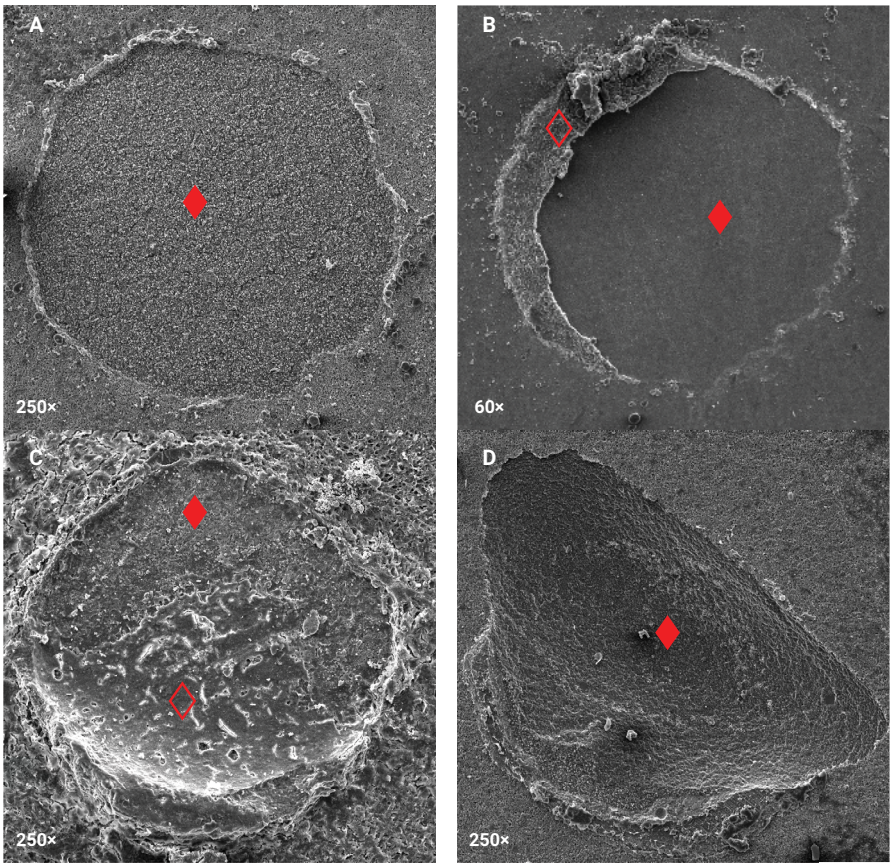

Figure 2. Representative micrographs (SEM images; 60-250× magnification) of the failure modes after the microshear test. A depicts the adhesive failure mode at the ceramic/cement interface; B demonstrates the predominantly adhesive failure mode; C exhibit a cohesive failure in cement and D shows a cohesive failure in ceramic. The symbol (♦) represents the ceramic surface free of cement, while the symbol (◊) represents the surface with the presence of resin cement.

Discussion

The specimens were subjected to thermocycling and water storage before the μSBS testing in order to age the adhesive interfaces between resin cements and the PICN material. Campos et al.[24] (2016), Lise et al.[26] (2017), Cekic-Nagas et al.[32] (2016) and Silva et al.[33] (2018) reported that the aging protocol drastically decreased the bond strength values compared to the baseline values. This occurrence would be due to the high molar concentration of the water and its small molecular size which allows its penetration in small spaces between polymer chains or functional groups, negatively affecting the thermal stability of the polymer, and in turn leading to its plasticization and hydrolytic degradation of resin cement[24,32,33]. Considering long-term values are more relevant, baseline data was not collected.

In this study it was observed that mechanical and chemical surface conditioning increased the μSBS values compared with CTRL; thus, the first hypothesis was accepted. In accordance with these results, Schwenter et al.[4] (2016) tested the shear bond strength of polished PICN and tree commercial resin cements and found that no or minor adhesion was obtained without silanization. This can be attributed to the industrial polymerization process of the polymer, with a high conversion degree of the monomers which limited the number of reactive vinyl groups available on the surface of the resin substrate and consequently a low amount of chemical bonding occurred[4]. Therefore, the bond durability between ceramic and resin cement needs to be ensured by surface treatments which increase the surface roughness[32]. The material's microstructure determines the surface treatment to be used. PICN has a dominant feldspathic ceramic network interpenetrated with minor polymer content. Thus, the surface treatment to achieve mechanical interlocking proposed herein was the same indicated for etchable ceramics (hydrofluoric acid etching) or for composite indirect restorations (sandblasting)[24].

HF promotes a selective dissolution of the glassy and crystalline phases of the restorative material[32]. Pre-treatment with 5% hydrofluoric acid for 60 seconds, as recommended by the manufacturer, promotes a two-fold increase in roughness than untreated ceramic, but statistically similar surface energy of the CTRL. Nevertheless, sandblasting showed an increase in roughness of around 6.8 times in relation to the untreated surface, and a meaningful enhancement in surface energy (smaller contact angle) (Table 3, Figure 1). The increase in surface roughness improves the interlocking between ceramic and resin cement[24]. However, sandblasting presents some limitations, as the possibility of ceramic surface contamination by sand particles and potential material damage, resulting in crack formation between the ceramic and polymer components and huge volume loss[20,34] which might be especially harmful in thinner pieces. Strasser et al.[20] (2018) showed that sandblasting with 50 μm/2 bar achieve an increase in roughness without harming the surface and might be comparable to HF at this point.

The findings of this study showed that the PICN surface treated with hydrofluoric acid etching and alumina sandblasting behaved similarly with respect to resin cement bonding; thus, the second hypothesis was also accepted. These findings are in agreement with Elsaka[11] (2014) who compared the microtensile bond strength of PICN

treated with HF and SB to resin cement before and after 30 days of water storage. However, it is important to note that the author used alumina particles (110 µm) and hydrofluoric acid concentrations (9%) which were different from those in this study (50 µm and 5%, respectively). Barutcigil et al.[10] (2019) also found similar behavior for both treatments, however the authors did not conduct aging procedures.

The third hypothesis was that the use of chemical conditionings following surface treatments of PICN would increase its bond strength to resin cement. This hypothesis was partially accepted, because only the groups that had silane application after HF etching and sandblasting (HF-S and SB-S) achieved higher bond strength to the resin cement than the mechanical conditioning alone (HF and SB). These findings are in agreement with Elsaka[11] (2014) and Lise et al.[26] (2017) who showed statistically similar µTBS to resin cements for sandblasted and etched PICN surfaces followed by silane application after water storage, respectively. Demirtag and Culhaoglu[35] (2019) also recommended silanization after both surface treatments and found slightly superior results for HF-S than SB-S. However, the authors performed the thermocycling of the specimens for 2000 cycles, while the thermocycling in the present study was conducted with 12000 cycles. Silane is a bifunctional molecule which has a silanol group that reacts with both the silica and integrated polymer components of hybrid ceramic surfaces and the methacrylate group that connects to the organic matrix of composites, improving bond strength of resin composites to ceramics[34]. Cohesive (in PICN) and predominantly adhesive fractures were the majority in relation to adhesive fractures in the silanized groups of this study (HF-S and SB-S). This type of failure was also found by Schwenter et al.[4] (2016), Cekig-Nagas et al.[32] (2016) and El-Damanhoury and Gaintantzopoulou[36] (2018). According to Elsaka[11] (2014) cohesive failure modes are preferable to fully adhesive failure, since adhesive failures are often associated with low bond strength values[11], which was also observed in the present study.

Clinical assessments also were conducted for PICN using the HF-S approach. In a three-year prospective clinical study of PICN single crowns adhesively cemented to dental abutments, Spitznagel et al.[37] (2020) founded a survival rate of about 93.9% and no debonding was observed. Oudkerk et al.[38] (2020) conducted a prospective clinical study of full-mouth rehabilitation of worn dentition (no-prep approach) using PICN restorations, finding that the survival rate of these restorations after 2 years was 100%, while the success rate was 93.5% due to presence of 11 minor chippings and one debonding.

The fourth hypothesis that the different chemical conditionings following surface treatments would have similar effects on the bond strength results was rejected, since the silanized groups (HF-S and SB-S) and the sandblasted group followed by universal adhesive (SB-UA) performed better than the other adhesive approaches (HF-S-A; HF-UA and SB-S-A).

The application of a bond agent after silanization without light curing was advocated by the International Academy for Adhesive Dentistry[7] for better penetration of the composite within the treated ceramic surface. The use of Single Bond 2 (3M ESPE) was an attempt to test this adhesive system which has the bonding agent and the primer in a single bottle (fifth generation adhesive)[39]. The groups with application of

this adhesive (HF-S-A and SB-S-A) showed intermediate bond strength values. However, the number of pre-test failures was quite significant, thus suggesting that this treatment is not reliable for the tested resin cement. It is important to note that this adhesive system is not the one recommended for combined use with RelyX Ultimate (3M ESPE) by the resin cement manufacturer[40]. Thus, further studies evaluating the combined use of adhesives and resin cements are necessary before contraindicating an adhesive system for conditioning PICN.

Single Bond Universal (3M ESPE) is an adhesive system which contains methacrylate-modified polyalkenoic acid copolymers, methacryloyloxydecyl dihydrogen phosphate (MDP) and silane[41]. Thus, in addition to the union of the silane agent to the ceramic component of PICN, acid groups of either the copolymer or MDP to urethane groups of the UDMA enabled bonding to the polymer part of the hybrid ceramic[22]. However, the group in which the Single Bond Universal was applied in addition to acid etching (HF-UA) in this study obtained intermediate microshear bond strength values and a high number of pre-test failures during thermocycling. In evaluating the bond strength of PICN with two types of resin cement after several acid etching protocols combined with silane, Single Bond Universal and an association of both primers, Rohr et al.[22] (2017) found that the highest bond strengths values were achieved when silane was applied followed by the universal adhesive after 5% hydrofluoric acid conditioning for 30 to 60 s. Corroborating the findings of the present study, the authors also did not find adequate mean bond strength values when universal adhesive was applied solely after HF[22]. Furthermore, Awad et al.[19] (2019) also found a significantly higher performance of silane group and low bond strengths of universal adhesives after aging in comparing the behavior of a silane primer, a silane-containing universal adhesive and a silane-free universal adhesive after HF etching (4.6%) on μTBS of PICN with resin cement. These results seem to demonstrate that chemical adhesion promoted by UA and PICN was not sufficient compared to silane (HF-S). This could be explained because the acidic pH of universal adhesive maintains the silanol as unstable, being subjected to hydrolysis and dehydration condensation, and is therefore less effective in forming a strong siloxane network[42]. Moreover, the monomers and hydrophilic solvents present in the adhesive composition might favor water sorption and plasticization of the adhesive interface. Without the adhesive layer, the hydrophobic resin cement achieves more adequate wetting on the etched PICN surface and an improved bond maturation occurs even after aging[43]. Additionally, the adhesive solution had a high viscosity compared to the silane-based primer, which may reduce its penetrative effects on the surface irregularities caused by acid etching[44].

Adhesive viscosity apparently did not have the same impact on the blasted PICN surfaces. The universal adhesive in the sandblasted group (SB-UA) behaved statistically similar to silanized groups (HF-S and SB-S), and seems to be a good alternative approach for hybrid ceramic pretreatment. Bayazit[12] (2019) also found the highest bond strength values to composite cement when the PICN was sandblasted followed by universal adhesive application compared to acid etching plus universal adhesive and control (no treatment), although an appropriate comparison cannot be conducted since the authors did not perform aging procedures.

However, Sagsoz et al.[23] (2019) found no statistical difference among shear bond strength of resin cement and PICN treated with HF, SB and CTRL followed by universal adhesive. In addition, no aging method was conducted and thus the long-term results cannot be accessed.

One of the limitations of this study was the expressive pretest failures in the CTRL, HF-S-A, HF-UA and SB-S-A groups. However, these were associated with lower microshear bond strengths and did not impair the statistical analysis, since at least one microcylinder remained per specimen for the mean calculation, except for the HF-S-A group. A second limitation of this study is the microshear testing approach which enables developing non-homogeneous stresses at the adhesive zone and interfaces, and may cause cohesive fractures in the materials. In fact, cohesive fractures in ceramic and in cement were found. Further studies are necessary to access the long-term bond strength of different types of resin cement to PICN, and other surface treatments and adhesive approaches can also be evaluated. Other studies should also be conducted to assess the bonding effect (provided by different protocols) on the fatigue strength of PICN in testing designs closer to clinical conditions.

Conclusion

1. The best bonding performances were achieved when the PICN surface was HF-etched followed by silanization, as recommended by the manufacturer, or sandblasted followed by silane or universal adhesive application.
2. The use of adhesive agents after hydrofluoric acid etching does not bring advantages to the bond between PICN and resin cement.
3. Sandblasting with alumina particles proved to be a good alternative approach to hydrofluoric etching.

Acknowledgments

The authors would like to thank Dental Cremer for supplying the materials for this research.

References

1. Coldea A, Swain MV, Thiel N. Mechanical properties of polymer-infiltrated-ceramic-network materials. Dent Mater. 2013 Apr;29(4):419-26. doi: 10.1016/j.dental.2013.01.002.
2. Awada A, Nathanson D. Mechanical properties of resin-ceramic CAD/CAM restorative materials. J Prosthet Dent. 2015 Oct;114(4):587-93. doi: 10.1016/j.prosdent.2015.04.016.
3. Mainjot AK, Dupont NM, Oudkerk JC, Dewael TY, Sadoun MJ. From artisanal to CAD-CAM blocks: state of the art of indirect composites. J Dent Res. 2016 May;95(5):487-95. doi: 10.1177/0022034516634286.
4. Schwenter J, Schmidli F, Weiger R, Fischer J. Adhesive bonding to polymer infiltrated ceramic. Dent Mater J. 2016;35(5):796-802. doi: 10.4012/dmj.2015-271.
5. Subaşi MG, Alp G. Repair bond strengths of non-aged and aged resin nanoceramics. J Adv Prosthodont. 2017 Oct;9(5):364-70. doi: 10.4047/jap.2017.9.5.364.

6. Vita Enamic®. Vita Enamic Working Instructions. [cited 2020 Jul 20]. Available from: https://www.vita-zahnfabrik.com/en/VITA-ENAMIC-24970.html.

7. Özcan M, Volpato CAM. Surface conditioning and bonding protocol for polymer-infiltrated ceramic: how and why? J Adhes Dent. 2016;18(2):174-5. doi: 10.3290/j.jad.a35979.

8. Facenda JC, Borba M, Corazza PH. A literature review on the new polymer-infiltrated ceramic-network material (PICN). J Esthet Restor Dent. 2018 Jul;30(4):281-6. doi: 10.1111/jerd.12370.

9. Spitznagel FA, Horvath SD, Guess PC, Blatz MB. Resin bond to indirect composite and new ceramic/polymer materials: a review of the literature. J Esthet Restor Dent. 2014;26(6):382-93. doi: 10.1111/jerd.12100.

10. Barutcigil K, Barutcigil C, Kul E, Özarslan MM, Buyukkaplan US. Effect of different surface treatments on bond strength of resin cement to a CAD/CAM restorative material. J Prosthodont. 2019 Jan;28(1):71-8. doi: 10.1111/jopr.12574.

11. Elsaka SE. Bond strength of novel CAD/CAM restorative materials to self-adhesive resin cement: the effect of surface treatments. J Adhes Dent. 2014 Dec;16(6):531-40. doi: 10.3290/j.jad.a33198.

12. Bayazıt EÖ. Microtensile bond strength of self-adhesive resin cements to CAD/CAM resin-matrix ceramics prepared with different surface treatments. Int J Prosthodont. 2019;32(32):433-8. doi: 10.11607/ijp.6268.

13. Yu H, Özcan M, Yoshida K, Cheng H, Sawase T. Bonding to industrial indirect composite blocks: a systematic review and meta-analysis. Dent Mater. 2020 Jan;36(1):119-34. doi: 10.1016/j.dental.2019.11.002.

14. Soares CJ, Giannini M, Oliveira MT, Paulillo LAMS, Martins LRM. Effect of surface treatments of laboratory-fabricated composites on the microtensile bond strength to a luting resin cement. J Appl Oral Sci. 2004 Mar;12(1):45-50. doi: 10.1590/s1678-77572004000100009.

15. Eldafrawy M, Ebroin MG, Gailly PA, Nguyen J-F, Sadoun MJ, Mainjot AK. Bonding to CAD-CAM composites: an interfacial fracture toughness approach. J Dent Res. 2018 Jan;97(1):60-7. doi: 10.1177/0022034517728714.

16. Emsermann I, Eggmann F, Krastl G, Weiger R, Amato J. Influence of pretreatment methods on the adhesion of composite and polymer infiltrated ceramic CAD-CAM blocks. J Adhes Dent. 2019;21(5):433-43. doi: 10.3290/j.jad.a43179.

17. Yoshida K, Kamada K, Atsuta M. Effects of two silane coupling agents, a bonding agent, and thermal cycling on the bond strength of a CAD/CAM composite material cemented with two resin luting agents. J Prosthet Dent. 2001 Feb;85(2):184-9. doi: 10.1067/mpr.2001.113628.

18. Matinlinna JP, Lassila LV, Özcan M, Yli-Urpo A, Vallittu PK. An introduction to silanes and their clinical applications in dentistry. Int J Prosthodont. 2004;17(2):155-64.

19. Awad MM, Albedaiwi L, Almahdy A, Khan R, Silikas N, Hatamleh MM, et al. Effect of universal adhesives on microtensile bond strength to hybrid ceramic. BMC Oral Health. 2019 Aug;19(1):178. doi: 10.1186/s12903-019-0865-7.

20. Strasser T, Preis V, Behr M, Rosentritt M. Roughness, surface energy and superficial damages of CAD/CAM materials after surface treatment. Clin Oral Invest. 2018 Nov;22(8):2787-97. doi: 10.1007/s00784-018-2365-6.

21. Alp G, Subaşi MG, Johnston WM, Yilmaz B. Effect of different resin cements and surface treatments on the shear bond strength of ceramic-glass polymer materials. J Prosthet Dent. 2018 Sep;120(3):454-61. doi: 10.1016/j.prosdent.2017.12.016.

22. Rohr N, Flury A. Fischer J. Efficacy of a universal adhesive in the bond strength of composite cements to polymer-infiltrated ceramic. J Adhes Dent. 2017;19(5):417-24. doi: 10.3290/j.jad.a39277.

23. Sagsoz O, Sagsoz NP, Yurtcan MT, Ozcelik N. Hydroxyapatite coating effect on the bond strength between CAD/ CAM materials and a resin cement. Odontology. 2019 Oct;107(4):491-9. doi: 10.1007/s10266-019-00420-y.

24. Campos F, Almeida CS, Rippe MP, de Melo RM, Valandro LF, Bottino MA. Resin bonding to a hybrid ceramic: effects of surface treatments and aging. Oper Dent. 2016;41(2):171-8. doi: 10.2341/15-057-L.

25. Mine A, Kabetani T, Kawaguchi-Uemura A, Higashi M, Yuko T, Hagino R, et al. Effectiveness of current adhesive systems when bonding to CAD/CAM indirect resin materials: A review of 32 publications. Jpn Dent Sci Rev. 2019 Nov;55(1):41-50. doi: 10.1016/j.jdsr.2018.10.001.

26. Lise DP, Van Ende A, De Munck J, Vieira LCC, Baratieri LN, Van Meerbeek B. Microtensile bond strength of composite cement to novel CAD/CAM materials as a function of surface treatment and aging. Oper Dent. 2017;42(1):73-81. doi: 10.2341/15-263-L.

27. Peumans M, Valjakova EB, De Munck J, Mishevska CB, Van Meerbeek B. Bonding effectiveness of luting composites to different CAD/CAM materials. J Adhes Dent. 2016;18(4):289-302. doi: 10.3290/j.jad.a36155.

28. Marchesi G, Frassetto A, Mazzoni A, Apolonio F, Diolosà M, Cadenaro M, et al. Adhesive performance of a multi-mode adhesive system: 1-year in vitro study. J Dent. 2014 May;42(5):603-12. doi: 10.1016/j.jdent.2013.12.008.

29. ISO 4287. Geometrical product specifications (GPS) – surface texture: profile method, terms definitions and surface texture parameters. Geneva: International Organization for Standardization; 1997.

30. Prochnow C, Venturini AB, Grasel R, Gundel A, Bottino MC, Valandro LF. Adhesion to a lithium disilicate glass ceramic etched with hydrofluoric acid at distinct concentrations. Braz Dent J. 2018 Sep-Oct;29(5):492-9. doi: 10.1590/0103-6440201802080.

31. Tedesco TK, Montagner AF, Skupien JA, Soares FZ, Susin AH, Rocha RO. Starch tubing: an alternative method to build up microshear bond test specimens. J Adhes Dent. 2013 Aug;15(4):311-5. doi: 10.3290/j.jad.a28602.

32. Cekic-Nagas I, Ergun G, Egilmez F, Vallittu PK, Lassila LVJ. Micro-shear bond strength of different resin cements to ceramic/glass-polymer CAD-CAM block materials. J Prosthodont Res. 2016 Oct;60(4):265-73. doi: 10.1016/j.jpor.2016.02.003.

33. Silva PNF, Martinelli-Lobo CM, Bottino MA, Melo RM, Valandro L.F. Bond strength between a polymer-infiltrated ceramic network and a composite for repair: effect of several ceramic surface treatments. Braz Oral Res. 2018;32:e28. doi: 10.1590/1807-3107bor-2018.vol32.0028.

34. Elsaka SE. Repair bond strength of resin composite to a novel CAD/CAM hybrid ceramic using different repair systems. Dent Mater J. 2015;34(2):161-7. doi: 10.4012/dmj.2014-159.

35. Demirtag Z, Culhaoglu AK. Surface roughness of ceramic-resin composites after femtosecond laser irradiation, sandblasting or acid etching and their bond strength with and without silanization to a resin cement. Oper Dent. 2019;44(2):156-67. doi: 10.2341/17-391-L.

36. El-Damanhoury HM, Gaintantzopoulou MD. Self-etching ceramic primer versus hydrofluoric acid etching: etching efficacy and bonding performance. J Prosthodont Res. 2018 Jan;62(1):75-83. doi: 10.1016/j.jpor.2017.06.002.

37. Spitznagel FA, Scholz KJ, Vach K, Gierthmuehlen PC. Monolithic polymer-infiltrated ceramic network CAD/CAM single crowns: three-year mid-term results of a prospective clinical study. Int J Prosthodont. 2020;33(2):160-8. doi: 10.11607/ijp.6548.

38. Oudkerk J, Eldafrawy M, Bekaert S, Grenade C, Vanheusden A, Mainjot A. The one-step no-prep approach for full-mouth rehabilitation of worn dentition using PICN CAD-CAM restorations: 2-yr results of a prospective clinical study. J Dent. 2020 Jan;92:103245. doi: 10.1016/j.jdent.2019.103245.

39. 3M ESPE. Adper™ Single Bond 2 Adhesive. Saint Paul: 3M Center; 2004 [2020 Jul 10]. Available from: https://multimedia.3m.com/mws/media/276868O/adper-single-bond-2-technical-profile.pdf.

40. 3M ESPE. RelyX™ Ultimate Adhesive Resin Cement. Technical Data Sheet. [cited 2020 Jul 10]. Available from: https://multimedia.3m.com/mws/media/783784O/3m-relyx-ultimate-adhesive-resin-cement-technical-data-sheet.pdf.

41. 3M Ciência Aplicada à vida™. Adesivo 3M Single Bond Universal. Saint Paul: 3M Oral Care; 2017 [cited 2020 Jul 10]. Available from: https://multimedia.3m.com/mws/media/1376776O/sbu-4-pages-brochure-pt-la.pdf.

42. Yoshihara K, Nagaoka N, Sonoda A, Maruo Y, Makita Y, Okihara T, et al. Effectiveness and stability of silane coupling agent incorporated in 'universal' adhesives. Dent Mater. 2016 Oct;32(10):1218-25. doi: 10.1016/j.dental.2016.07.002.

43. Romanini-Junior JC, Kumagai RY, Ortega LF, Rodrigues JA, Cassoni A, Hirata R, et al. Adhesive/silane application effects on bond strength durability to a lithium disilicate ceramic. J Esthet Restor Dent. 2018 Jul;30(4):346-51. doi: 10.1111/jerd.12387.

44. Alrahlah A, Awad MM, Vohra F, Al-Mudahi A, Al jeaidi ZA, Elsharawy M. Effect of self etching ceramic primer and universal adhesive on bond strength of lithium disilicate ceramic. J Adhes Sci Technol. 2017;31(23):2611–9. doi: 10.1080/01694243.2017.1312079.