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Original Article

Bond strength of self-adhesive resin cements to a high transparency zirconia crown and dentin

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KEYWORDS
Self-adhesive resin cement;
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Surface treatment;
Shear bond strength

Abstract *Background/purpose:* The bond strength and durability of highly translucent zirconia ceramics to dentin is still unclear. The purpose of this study was to investigate the effect of various surface treatments on the bond strength of self-adhesive resin cements to high-translucent zirconia crowns and dentin.
Materials and methods: A high-transparent zirconia and three self-adhesive resin cements (G-CEM LinkAce (GCL), RelyX U200 (RXU) and TotalCem (TTC)) were used. The zirconia surface was sandblasted with 50 μm alumina particles or coated with an SR Link primer, while a dentin primer (Tetric N-Bond Universal, TBU) was applied to the surface of the dentin. By using three self-adhesive resin cements, zirconia samples were bonded to the dentin surfaces of human teeth. The shear strength of the specimens was measured before and after 10,000-cycle thermocycling or 90-day aging.
Results: When using GCL to bond with the untreated dentin and various zirconia surfaces, the shear bond strength of the sandblasted (Z_{SB}) and RS Link primer-coated (Z_{LK}) groups was significantly higher than that of the untreated control group (Zc). However, in the case of TBU-treated dentin, the shear strength of the $Z_{SB\ +\ LK}$ + D_{TBU} group was significantly higher than

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that of the other groups. After thermocycling and aging, the shear strength of the $Z_{SB+LK}+D_{TBU}$ group using GCL and RXU cements decreased slightly, while the TTC showed no impact.

Conclusion: The zirconia surface pretreated by sandblasting and bonding agent, which was sequentially bonded with a primer-treated dentin by using resin cements, can provide excellent shear bond strength and anti-aging performance.

Introduction

Because of the superior chemical stability, biocompatibility, and supreme mechanical properties among dental ceramics, zirconia has been widely used as a biomaterial for implants and all-ceramic dental restorations such as cores, framework, post and inlays.[1–3] With the increasing demand of patient for aesthetic considerations of dental restorations, numerous manufacturers have launched new zirconia ceramic materials with optical transparency similar to natural teeth, which are yttria-stabilized tetragonal zirconia polycrystal ceramics (Y-TZP) ceramics with different formulations (3Y-TZP -, 4Y- TZP and 5Y-TZP).[4,5] By increasing the content of yttrium oxide, more cubic phases are generated, thereby improving the translucency of zirconia ceramics[1,6] that can serve as alternatives to conventional glass-ceramic materials for aesthetic rehabilitations, including dental veneers, onlays, crowns and bridges.

Dental cements are usually used to bond the restoration to the abutment teeth in crown and bridge restorations. However, the composition of the dentin is more complex than that of the enamel, and it is less calcified structure and contains hydrophilic collagen fibers.[7] The openings of dentin tubules on the surface of the dentin make the dentin a highly permeable and high-moisture tissue, which may be adversely to apply the dental adhesion system on the dentin surface.[8] In addition, the smear layer on the tooth surface after clinical tooth preparation is also a factor that is not conducive to bonding. The interfacial bonding of dental cement to the surface of tooth and to dental restorations is the most crucial factor that is related not only to the long-term function of the restorations, but also related to the lifespan of the teeth.

Self-adhesive resin cements have been clinically used for nearly 20 years. It aims to simplify the operation of conventional resin cement pretreatment, prevent saliva and blood contamination during the complicated resin cement attachment operation, reduce operator error, shorten the patient's chair time, and decrease the treatment burden. Since the self-adhesive resin cement itself contains an acidic monomer with acid etching function, there is no need to perform a complicated bonding procedure (multi-step) on the surface of the tooth before bonding, which simplifies pretreatment compared with conventional resin cement.[9] At present, there are many kinds of commercial self-adhesive resin adhesives. In the basic composition, in addition to the main resin matrix (e.g., Bis-GMA, UDMA and TEGDMA), they each contain different multifunctional monomers with adhesive properties, such as hydroxyethyl methacrylate (HEMA), 4-methacryloxyethylenetrimellitic anhydride (4-META), 10-methacryloyloxydecamethylene phosphate (MDP), dimethacrylate and phosphoric acid ester monomer. The different binding stability of these monomers and hydroxyapatite results in the differences in the bond strength between self-adhesive resin cements of different brands and zirconia or teeth tissue.[10] It is worth noting that the filler content and particle size in the cement, and even the acid-base pH value are also different, which could affect the bond strength between the self-adhesive resin adhesive and zirconia or teeth.[11] However, the bonding strength and durability between zirconia, resin cement and tooth matrix do not seem to be satisfactory,[12] because the resin bonding agent absorbs water, causing dentin bonding to fail.[13]

Given that zirconia ceramics are known to have exception chemical stability, their low surface reactivity may limit the bonding efficiency between zirconia and resin cement.[14,15] To solve the poor bonding between zirconia and resin cement, some studies have been conducted to change the surface structure or chemical properties of zirconia through alumina blasting,[16,17] tribochemical silica coating,[18] selective infiltration etching[19] and laser treatments.[17] It is also recommended to use a primer on zirconia surfaces to enhance the bonding strength of the resin and zirconia.[18] According to the manufacturer's recommendation, SR Link is a metal or zirconia/composite bonding agent that provides a covalent bond between metal or zirconia frameworks and composite resin. However, there is little information on the bond strength of the SR Link applied to zirconia materials. In addition, there have been few studies on resin cements for dental crowns and bridges with high-transparency zirconia ceramics for aesthetic rehabilitations, and further research is needed.

The aim of the present study was to study effect of surface treatments (sandblasting or primer) on bond strength of highly transparent zirconia to dentin bonded with self-adhesive resin cements. The bond strength durability of the zirconia-teeth assembly was also evaluated before and after thermocycling and aging.

Table 1 Materials used in this study.

Material	Brand (Code)	Composition (%)	Lot. No.	Manufacturer
Zirconia	NexxZr T	ZrO_2: ≥89%; Y_2O_3 : 4—6%; HfO_2: ≤5%; Al_2O_3: <1	TAAMSR	Sagemax Bioceramics Inc., Federal Way, WA, USA
Self-adhesive resin cement	G-CEM LinkAce (GCL)	Phosphonate monomer, methacryloxypropyl-trimethoxysilane, dimethylbenzyl hydroperoxide, fluoro aluminosilicate glass, silicon dioxide, initiator, inhibitor, pigments.	1,902,281	GC Corporation, Tokyo, Japan
	RelyX U200 (RXU)	Base paste: methacrylate monomers containing phosphoric acid groups, methacrylate monomers, silanated fillers, initiator components, stabilizers, rheological additives. Catalyst paste: methacrylate monomers, alkaline fillers, silanated fillers, initiator components, stabilizers, rheological additives, Pigments.	6,560,749	3 M ESPE, Neuss, Germany
	TotalCem (TTC)	UDMA, Bis-GMA, TEGDMA, 4-methacryloxyethyltrimellitic acid, barium glass, fumed silica.	4265-35HQBSEETR	Itena Clinical, Paris, France
Bonding agent	SR Link (LK)	Dimethacrylate, phosphate ester, solvents and benzoyl peroxide.	Y47445	Ivoclar Vivadent AG, Schaan, Liechtenstein
Dentin primer	Tetric N-Bond Universal (TBU)	Methacrylates, ethanol, water, highly dispersed silicon dioxide, Initiators and stabilizers	K50984	Ivoclar Vivadent AG

Materials and methods

Specimen preparation

A commercially available high-transparency 42% zirconia ceramics for CAD/CAM (NexxZr T, Sagemax bioceramics Inc., Federal way, WA, USA) was used as the full ceramic crown material to fabricate a square-shaped specimen with a dental model saw (G2 Concept, Schick dental Geräte, Schemmerhofen, Germany) (Table 1). The surface of the specimen was polished by using #800 SiC abrasive paper, then ultrasonically cleaned in distilled water for 10 min, and air-dried. According to the manufacturer's recommendation, the specimen was sintered from room temperature to 1300 °C at a heating rate of 30 °C/min and continued to 1530 °C at 40 °C/min for holding 2 h, and then cooled to 900 °C at 15 °C/min and room temperature at 20 °C/min using a sintering furnace (Vario S400, Zubler USA, Dallas, TX, USA). The dimension of the final zirconia specimen was 10 mm in length, 5 mm in width and 3 mm in thickness.

Dentin specimens were made from extracted human maxillary or mandibular first and second molars. First, the teeth were sterilized and soaked in 0.4% thymol solution. The root of a cavity-free human tooth was embedded in epoxy resin, and then a rectangular dentin specimen with a flat exposed dentin surface was cut on the buccal or lingual side of the crown using a slow-speed cutter (CLM40, Precision tabletop cutting machine, Power assist instrument scientific Corp., Taoyuan, Taiwan). The square dentin specimen was polished by using #600 SiC sandpaper to 4 mm in length, 4 mm in width and 2 mm in thickness. The dentin specimens were ultrasonically cleaned in 99.5% alcohol and deionized water for 5 min, respectively. The prepared teeth specimens were stored in water at 4 °C until use.

Surface treatment

Surface treatments of the zirconia specimens were performed by sandblasting (Z_{SB}), coating a SR Link primer (Z_{LK}) layer, and combining sandblasting and primer coating. The design of various specimen codes is shown in Fig. 1. For SB, the zirconia surface was sandblasted with 50 μm Al_2O_3 for 15 s from a distance of 15 mm under a pressure of 0.3 MPa. In the case of LK coating, the zirconia surface was coated

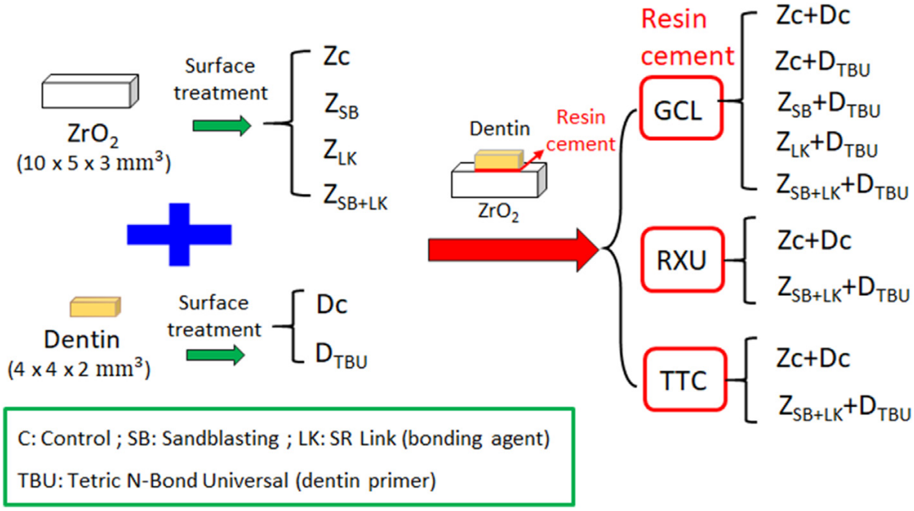

Figure 1 Diagram of the experimental groups in this study.

with a layer of SR Link bonding agent using a clean disposable brush and allowed to react for 3 min. For the dual treatment, the SB-treated zirconia surface was sequentially coated with LK (Z_{SB+LK}).

For the surface treatment of the dentin specimen, according to the manufacturer's instructions, it was coated with a universal TBU thin layer (Table 1) for 15 s using a small brush. After rinsing with water for 5 s and dried with paper, a light curing machine LED (Ultralite 1000 E, Rolence, Taoyuan, Taiwan) was used for 10 s of light curing.

On the other hand, for the zirconia-cement-dentin bonding test, the group in which both the zirconia and the dentin surface were not treated was regarded as the control group (Zc + Dc) (Fig. 1). In the experimental groups, a resin cement was used to bond the treated zirconia specimen to the TBU-treated dentin (D_{TBU}).

Self-adhesive resin cement bonding to zirconia

In order to ensure the actual adhesion area, a transparent tape layer with preformed holes of 4 mm in diameter and 0.06 mm in thickness was placed on the surface of the zirconia specimens with different treatments. According to the manufacturer's instructions, three different commercially self-adhesive resin cements (Table 1) were mixed for 20 s and then applied to the prepared space on the zirconia surface to make the zirconia specimens adhere to the dentin specimens with and without TBU primer. After that, a load of 650 g was immediately applied to the dentin for 30 s,[20–22] and then the excess resin cements were removed from the surrounding area. The dental LED light curing machine (Ultralite 1000 E, Rolence, Taoyuan, Taiwan) was used to irradiate the specimens for 20 s on each side that was a total of 80 s. All specimens were stored in deionized water at 37 °C for 24 h before shear bond test.

Surface and interface morphology

The surface morphology of all treated specimens and interface morphology of resin cement-zirconia or resin cement-dentin specimens were observed by field emission scanning electron microscope (FESEM; JSM-6700 F, JEOL, Tokyo, Japan).

Shear bond strength

The EZ Test machine (EZ-SX, Shimadzu, Kyoto, Japan) with a crosshead rate of 1.0 mm/min was used to perform the shear test. The ten specimens were measured for each condition ($n = 10$). The shear bond strength of the specimens was calculated using the following formula.

$F=P/A$, F = shear bond strength (MPa); P = load at fracture (N); S = area (mm^2).

Due to human dietary needs, the temperature and pH value in the mouth will change. Furthermore, the physiological moist environment in the mouth and the effect of saliva may cause the cement material to dissolve, which in turn reduces the ability to resist microleakage. Finally, the debonding of the prosthesis is detrimental to its durability and service life. For the durability of the zirconia ceramic-resin cement-dentin interface, the aqueous aging test is also one of the most commonly used evaluation methods. Different aging times[23] can be used to evaluate the anti-aging properties of various materials. However, from the perspective of clinical practice, a long-term period of time, such as 90 days, may be used to evaluate the effects of aging. In this study, to simulate the thermal change in the oral environment, the specimens were repeatedly soaked in a deionized water bath of 5 and 55 °C for a dwell time of 30 s using a thermal cycle device (TBN-971105, Ten billion, Tainan, Taiwan). On the other hand, the specimens were immersed in deionized water at 37 °C for 90 days of aging. After 10,000 cycles or aging tests, ten measurements of the specimens were made in the control group and the $Z_{SB+LK} + D_{TBU}$ group.

Fractured surface and fracture mode analysis

The fractured surface between zirconia and dentin was examined after the shear test using an optical microscope to evaluate the fracture mode of the specimens, in addition

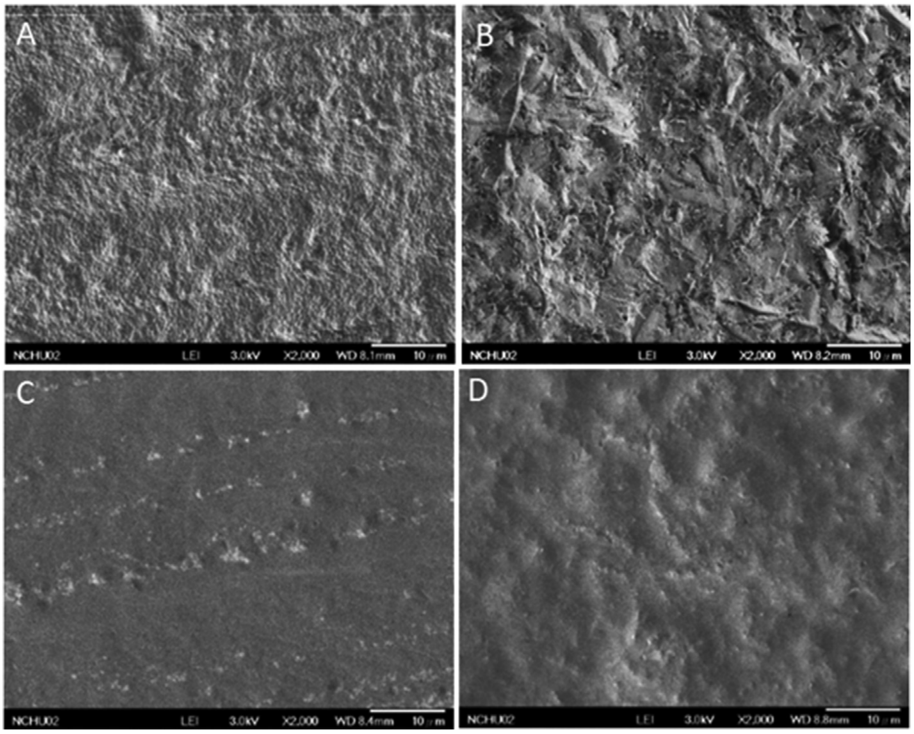

Figure 2 SEM images of zirconia specimens with and without surface treatment. (A): Without surface treatment; (B) Sandblasting; (C) Coated with SR link; (D) Combining sandblasting and SR link coating. Original magnification ×2000.

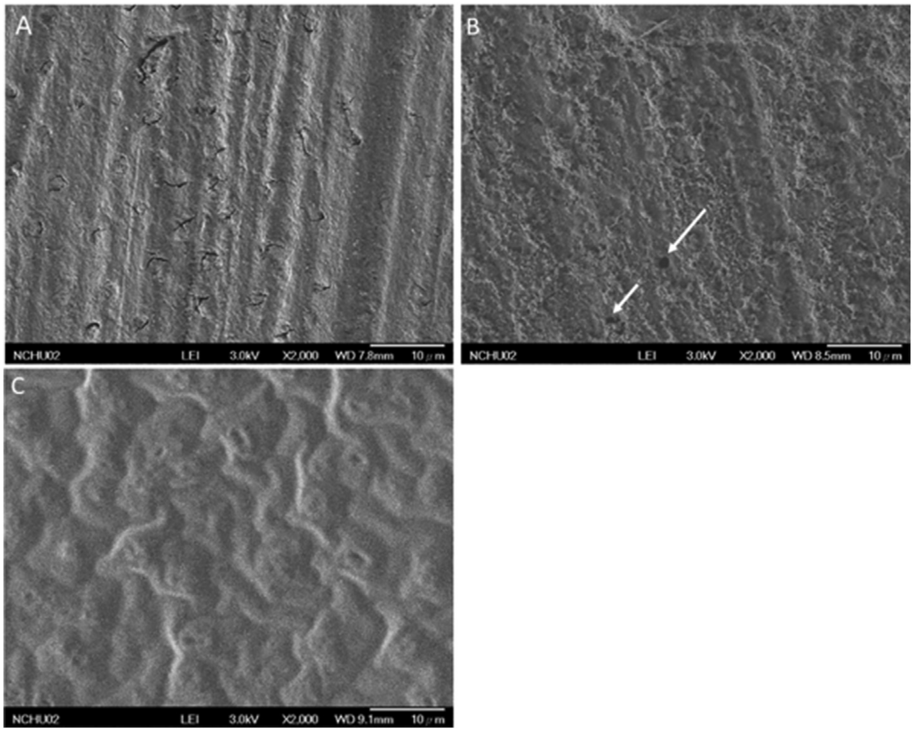

Figure 3 SEM images of dentin specimens with and without surface treatment. (A) Without surface treatment; (B) Applying TBU primer before curing; (C) Applying TBU primer and curing. The arrow indicated the opening of the dentin tubule. Original magnification ×2000.

to the fracture observation of FESEM. The failure mode was evaluated by scoring A, B, C and D. A: Adhesive failure on the dentin surface; B: Mixed failure at the dentin-resin cement interface; C: Adhesive failure on the zirconia surface; D: Mixed failure at the zirconia-resin cement interface.

Statistical analysis

One-way analysis of variance (ANOVA) and Tukey—Kramer comparison were used for statistical evaluation of shear bond strength. Two-way analysis of variance and Tukey—Kramer comparison were used for the statistical evaluation of shear bond strength in resin cements before and after the aging thermocycling. Statistical calculation was performed using statistics software (JMP 14, SAS Institute Inc., Cary, NC, USA). The significant was set as 0.05.

Results

Surface morphology of treated zirconia and dentin

SEM analysis shows that the surface morphology of the pretreated zirconia (Fig. 2) was completely different from that of the control group. Compared with the untreated surface (Fig. 2A), the sandblasting-treated zirconia appeared more irregular surface (Fig. 2B). In the SR link coating group, the surface becomes flatter (Fig. 2C), because some grains and grain boundaries disappeared as a result of the presence of SR link coating on the specimen surface. The surface undulation and roughness of the SB + LK group were somewhat increased compared with the single coating group (Fig. 2D).

Regarding the dentin specimen (Fig. 3), a relatively dense smear layer structure was observed to cover the untreated surface of the dentin specimen (Fig. 3A). Cracks

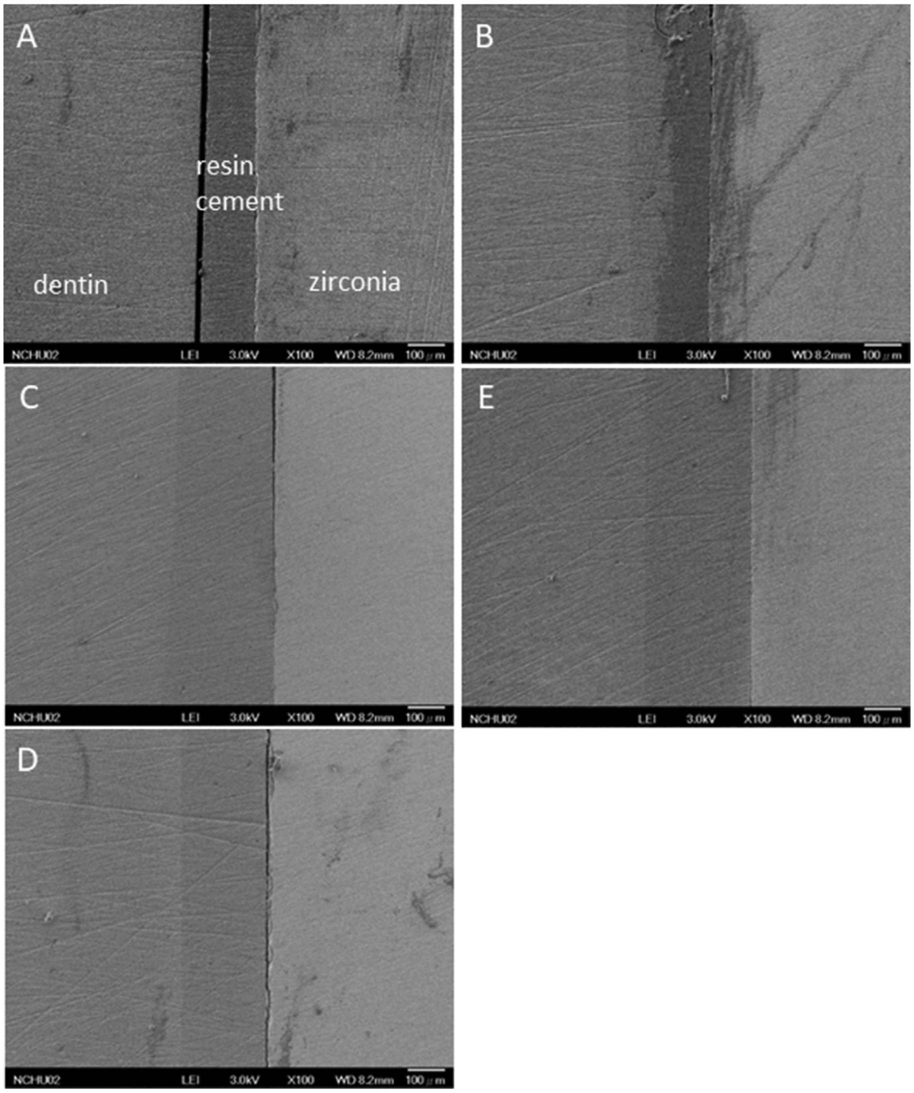

Figure 4 Cross-sectional SEM images of GCL resin cements bonded with various treated dentin and zirconia. (A): No surface treatment (Zc + Dc group); (B): Only on the dentin coated with TBU primer (Zc + D_{TBU} group); (C): Sandblasting zirconia and dentin coated with TBU primer (Z_{SB} + D_{TBU} group); (D): SR link-coated zirconia and dentin coated with TBU primer (Z_{LK} + D_{TBU} group); (E): Dual sandblasted and SR link-coated zirconia and dentin coated with TBU primer ($Z_{SB\ +\ LK}$ + D_{TBU} group).

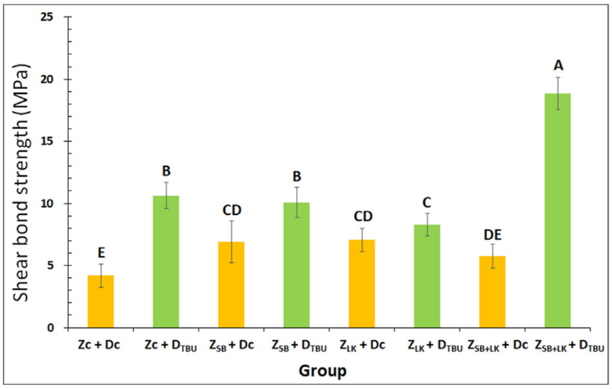

Figure 5 The shear bond strength of GCL resin cement to dentin with and without TBU treatment and zirconia with and without various treatments. The same letter means no significant difference ($P > 0.05$).

in the smear layer may be the openings of the dentin tubules. Compared with untreatment, only a few tubular openings of dentin were observed on the dentin surface after TBU primer treatment, as well as porous and reticulated smear layers (Fig. 3B). After TBU primer curing (Fig. 3C), the dentin surface was completely covered by the primer coating.

Cross-sectional structure

Taking GCL resin cement as an example, the cross-sectional images of the dentin-resin cement-zirconia specimens with different treatments are shown in Fig. 4. The resin cement layer attached well to the dentin of the experimental group, except for the control group (Fig. 4A). Debonding was found at the resin cement—zirconia junction in the $Z_{SB} + D_{TBU}$ (Fig. 4B), $Z_{SB} + D_{TBU}$ (Fig. 4C) and $Z_{LK} + D_{TBU}$ groups (Fig. 4D). It is worth noting that compared with other groups, the $Z_{SB + LK} + D_{TBU}$ group showed very good adhesion on both interfaces (Fig. 4E). No hybrid layer was seen at the interface of dentin and resin cement, and the cement was firmly bonded to the dentin.

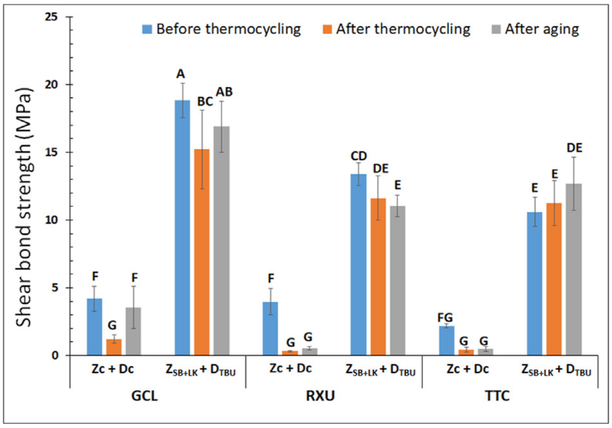

Figure 6 In terms of three self-adhesive resin cements, the shear bond strength of the specimens before and after thermocycling and aging in control and $Z_{SB + LK} + D_{TBU}$ groups. The same letter means no significant differences ($P > 0.05$).

Shear bond strength

Fig. 5 shows the shear bond strength of GCL self-adhesive resin cement to pretreated dentin and zirconia. The pretreated zirconia specimen demonstrated significantly superior bond strength compared with the control group ($P < 0.05$), regardless of the treatment method. The results also demonstrated that the bond strength of the specimens with different treatments on both the zirconia and the dentin surfaces was significantly higher than that of the control group ($Zc + Dc$ group) and the groups of treated zirconia specimens alone ($Z_{SB} + Dc$, $Z_{LK} + Dc$ and $Z_{SB + LK} + Dc$ groups) ($P < 0.05$), except for the $Z_{LK} + D_{TBU}$ group. The strength of the GCL specimens in the $Z_{SB + LK} + D_{TBU}$ group was the highest value (18.8 MPa) among the experiment groups, indicating a significant difference ($P < 0.05$).

Regarding the effects of thermocycling or aging, the shear bond strength of the three resin cements used to bond zirconia and dentin are shown in Fig. 6. After aging test, the strength of all control groups exhibited very low bonding values ranging from 0.3 to 3.6 MPa. In the experimental groups with pretreated surface, the strength of GCL and RXU tended to decrease after thermocycling. In contrast, the strength of the TTC group increased slightly, and the difference before and after thermocycling was not significant ($P > 0.05$). Furthermore, in the results of aging, the bond strength between zirconia and dentin varied with the brand of resin cement. There was a significant decrease in the strength of the RXU group ($P < 0.05$), a slight decrease in the GCL group, and a significant increase in the TTC group ($P < 0.05$). According to the 2 way ANOVA statistical analysis, the bond strength of the pretreated specimens with the GCL resin cement was significantly stronger than the other two brands of cements regardless of before and after the thermocycling and aging test ($P < 0.05$). The brand of self-adhesive resin cement and experimental conditions were the factors that affected the bond strength between zirconia and dentin ($P < 0.05$), and these two factors also had an interactive effect on the bond strength ($P < 0.05$). Furthermore, compared with the control group, the strength of the experimental group had a smaller decrease in strength after the aging experiment, regardless of the brand of resin cement.

Fracture mode

After the shear test, the amount of resin cement residue was observed on the zirconia surface or on the dentin surface and calculated. Fracture modes (%) of the specimens on the zirconia and dentin surfaces were visualized by using optical microscope in Fig. 7 and Fig. 8. For the control groups ($Zc + Dc$ group) of three self-adhesive resin cements, regardless of the brands of resin cement or experimental conditions, their most frequent fracture mode occurred at the dentin-resin cement or zirconia-resin cement interface (Fig. 7 A, B and C). Compared with the control group, the $Z_{SB + LK} + D_{TBU}$ groups showed few adhesive fractures in the specimens without aging test, while most of them showed mixed fractures regardless of the cement used. Moreover, especially after thermocycling and

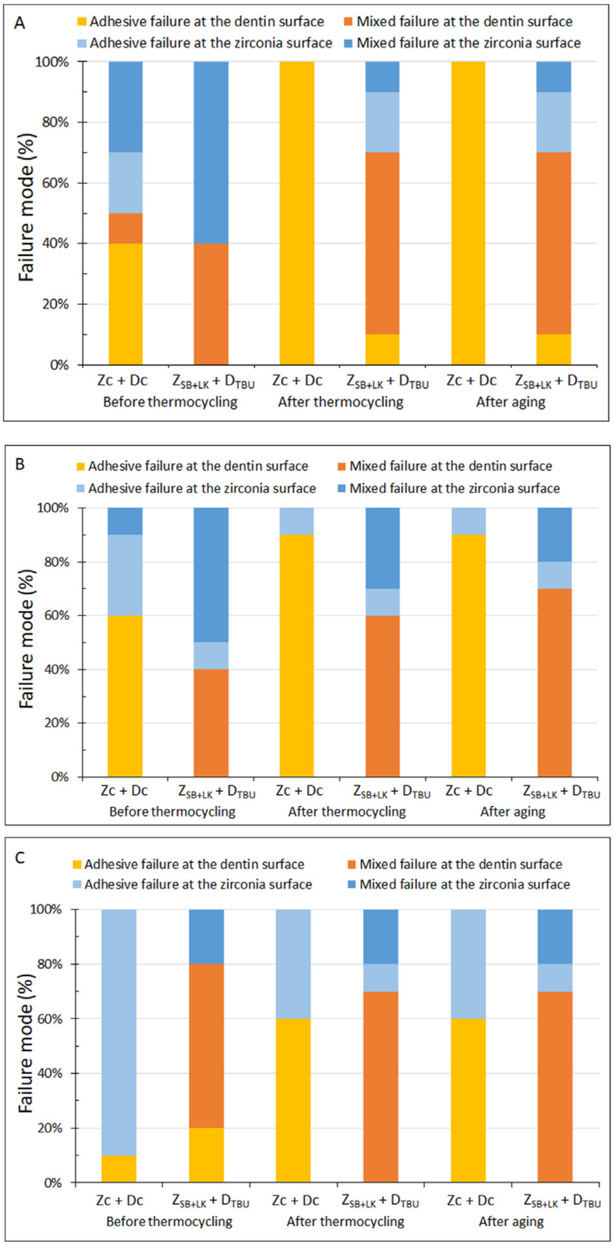

Figure 7 Fracture modes (%) of zirconia and dentin specimens after shear test. A, B, and C: GCL, RXU and TTC resin cement before and after aging test, respectively.

aging, the fracture mode of the experiment groups was still mostly mixed fracture mode.

Discussion

Zirconia ceramics do not contain a glass phase (silica) and have a relatively non-polar surface with extremely high chemical stability.[24,25] The inherent roughness of the surface of zirconia crowns made with CAD/CAM equipment is low, which is not sufficient to provide micromechanical inlay force, so it is not easy to obtain ideal bonding results with dental cements.[26,27] The results of the present study confirmed that the shear strength value of the pretreated zirconia specimens was higher than that of the control group, when GCL resin cement was used. Using GCL resin cement, the shear strength of the sandblasting the or primed group alone were 6.9 and 7.1 MPa, respectively, which was 1.5 times the bond strength of the untreated control group of 4.2 MPa. The reason should be attributed to the fact that sandblasting treatment can increase the surface area and roughness of zirconia and enhance the mechanical locking effect.[27–30] SEM observation of the surface morphology of the SB group samples confirmed this point, consistent with the previous study.[31] Kern et al. studied the surface roughness and tensile strength of zirconia treated by sandblasting with 50 μm alumina under a pressure of 0.05–0.25 MPa, and found that the tensile strength between Multilink Automix composite luting resin and zirconia increased.[28] In this study, the surface roughness obtained by the sandblasting conditions with zirconia would affect the bond strength between high-translucent zirconia and the resin cement.

The SR link coating treatment can use the adhesion monomer in the primer containing carboxylic acid and phosphoric acid functional groups to chemically bond the functional groups of zirconia or resin materials, thereby improving the adhesion to the surface of the zirconia.[9] This means that surface pretreatment with adhesion monomers was indispensable for zirconia surfaces. On the other hand, the combination of sandblasting and primer treatment on the zirconia surface has been proven to be effective in improving zirconia and dentin.[28,32,33] The fractures in the Z_{SB} + Dc, Z_{LK} + Dc and $Z_{SB + LK}$ + Dc groups all occurred on the dentin surface and showed adhesive fractures. The present study of zirconia specimens after sandblasting or coating treatment was effective in improving the bond strength between zirconia ceramics and dentin, which was consistent with the results of previous studies.[26,31] The results of this study also proposed that before the use of self-adhesive resin cements, zirconia should be pretreated by sandblasting or applying a bonding agent.[9]

When a self-adhesive resin cement was used, only relying on the treatment on the surface of the zirconia will be limited in improving the bond strength of zirconia to the dentin. In the results of this study, the shear strength value of the dentin surface treated with the TBU primer substantially increased compared to the shear strength of the untreated dentin, regardless of the type of self-adhesive resin cements used. The reason for this should be attributed to the presence of hydrophilic monomers (hydroxyethyl methacrylate/HEMA) and hydrophobic monomers (decanediol dimethacrylate/D_3MA) in the composition of the TBU primer with low acidic monomer content, resulting in a pH of about 2.5–3.0 for the TBU primer itself, which has a mild etching effect on the surface of the tooth.[34] This was confirmed by SEM, which revealed that the original smear layer on the surface of the tooth after TBU primer treatment was acid-etched into an existing porous shape. It can also be observed in the SEM cross-sectional view that all dentin specimens treated with TBU had a dense bond at the dentin-resin cement interface. Furthermore, since TBU also contains hydrophilic phosphate groups of MDP, it can promote mild acid demineralization and chemical bonding with hydroxyapatite. In addition, hydrophilic HEMA can promote penetration of dry and moist dentin through mild etching.[35]

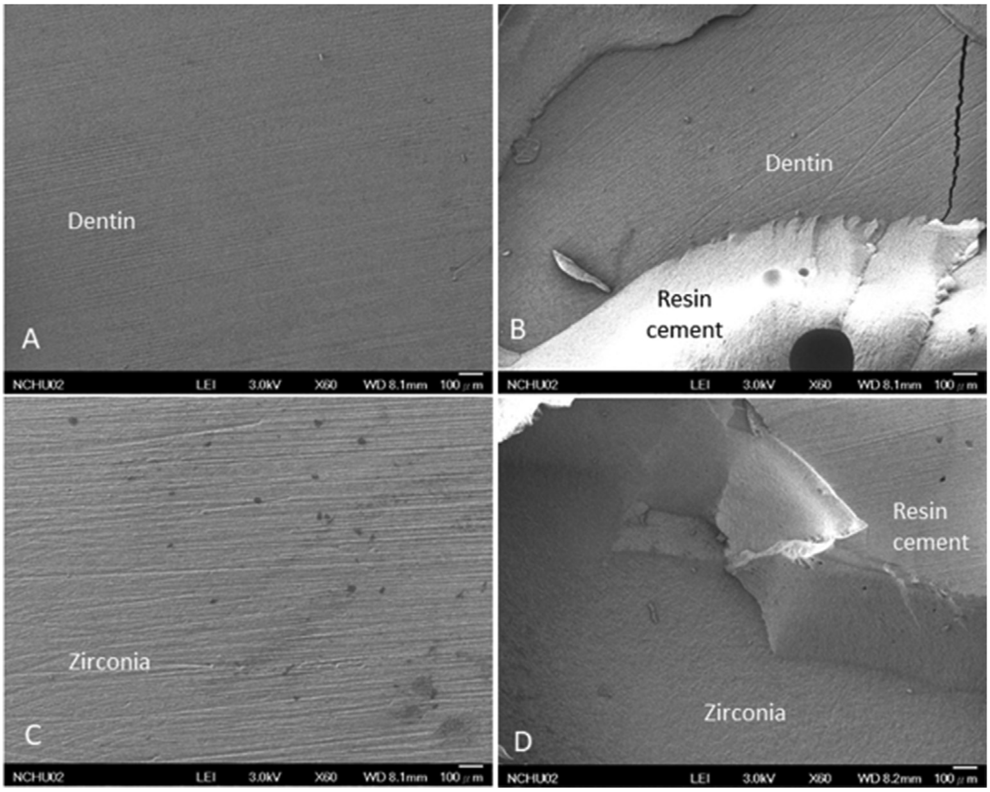

Figure 8 The fracture mode occurred at the dentin-resin cement or zirconia-resin cement interface. (A) Adhesive failure on the dentin surface; (B) Mixed failure at the dentin-resin cement interface; (C) Adhesive failure on the zirconia surface; (D) Mixed failure at the zirconia-resin cement interface.

MDP and MCAP adhesion monomers can maximize the contact between the adhesive and the dentin surface in a limited time of 15 s.[36–38] Therefore, TBU plays an important role as a bridge between the hydrophilic tooth surface and the hydrophobic resin restorative material to form a more reliable bond with the self-adhesive resin cement. As expected, the use of a coating treatment with enhanced tooth surface adhesion can significantly increase the bond strength of zirconia to dentin. The reason was that the adhesive monomer in the resin cements may spread more easily to the surface of the demineralized dentin after using TBU to partially remove the smear layer, thus improving the bond strength of the resin adhesive to the dentin. These facts were also confirmed by the fact that most of the fracture modes of the $Z_{SB\ +\ LK} + D_{TUB}$ group were mixed fracture modes.

The chemical composition, characteristics, and mechanical properties of resin cements may also affect the bond strength to zirconia.[32,39,40] In this study, the highest shear bond strength values of the three self-adhesive resin cements were GCL, followed by RXU; TTC showed the lowest SBS results. The reason may be the presence of phosphate monomers or phosphate functional groups in the two GCL and RXU cements. The two cements form a relatively strong and stable bond with zirconia through the chemical bond on the phosphoric acid ester monomer, which ensures a certain degree of bond strength with zirconia.[41,42] Moreover, 4-META in the TTC resin matrix has an acidic adhesion monomer, which can cause the surface of the dentin to be etched by acid. Its methacrylate group can be copolymerized with resin-based methacrylate monomers (TEGDMA, Bis-GMA, and UDMA). Furthermore, 4-META has an affinity for dental structure and promotes penetration of matrix monomer into dental hard tissue to improve adhesion between resin cement and restoration and tooth. The low bond strength of TTC may be due to the fact that the filler of TTC is mineral nanoparticles,[41,43] which has a slightly lower filler content in cement than that of GCL (60–70%)[44] and RXU (70%).[11]

According to the literature,[11,45,46] the shear bond strength between zirconia and dentin using self-adhesive resin cement is relatively weak. In order to improve the adhesion between self-adhesive resin cement and the dentin, proper pretreatment of dentin is required.[47] Although compared with total etching, the use of TBU primer to remove the smear layer and demineralize the dentin is not sufficient to form a hybrid layer.[9] However, the current results demonstrated that pretreatment of dentin with TBU alone can enhance the bond strength of the dentin to zirconia. Compared with using either sandblasting or primer treatment of zirconia, the synergistic efficacy of the dual treatment of sandblasting and primer coating on the zirconia surface and TBU coating on dentin can significantly enhance the bond strength between zirconia ceramics and human dentin, which is an effective, simple, and feasible surface modification method.

To simulate the oral environment, 10,000-cycle thermocycling and 90-day aging were used to evaluate the bond durability of three self-adhesive resin cements to zirconia and dentin. As a result, the pretreated specimens in the GCL and RXU groups had a smaller decrease in strength after aging, compared with the control group. The bond strength of the pretreated specimens in GCL and RXU was between 10.6 and 16.9 MPa, retaining 80—100% of the original strength that revealed good adhesion stability. The decrease in bond strength after artificial aging may be due to the difference in the thermal expansion coefficient between dentin-resin and cement-zirconia materials.[48] In addition, the functional monomers and hydrophilic functional groups in the resin cement may lead to hydrolysis.[49,50] After artificial aging and thermocycling, the failure mode of the experimental group was mainly mixed failure, but the control group developed adhesive failure at either the zirconia-resin cement interface or the dentin-resin cement interface due to the lack of primer and sandblasting treatment.

Additionally, it should be noted that although the pretreated specimens in the TTC group indicated the lowest strength among the three resin cements, these values did not decrease and remained stable after 90 days of water storage and thermocycling. This indicated that TTC had good bond stability. This is due to the mineral nanoparticle filler in TTC and the good affinity of 4-META adhesion with tooth structure, which promotes the penetration of matrix monomers into the dentin, forming a copolymer with low water absorption and low water solubility.[51]

In summary, the present results showed that the bond strength of the various pretreated specimens was greater than that of the control specimens with untreated surfaces. In particular, the SB/LK-treated zirconia and TBU-coated dentin specimens exhibited superior bond strength, regardless of the brand of the resin cement. The failure mode of the pretreated zirconia and dentin specimens bonded with the three resin adhesives was dominated by mixed failure, no matter what kind of aging and thermocycling tests they were underwent to. The experimental results proved that if the surfaces of zirconia and dentin were not treated, the bond strength of self-adhesive resin cements to high-translucent zirconia and dentin would be very low. In contrast, the bond strength of resin adhesives could be greatly improved by different surface pretreatments. In order to ensure the bonding effect of zirconia all-ceramic crowns, it is necessary to establish a good mechanical inlay and chemical bond between the zirconia restoration and the dentin through surface treatment, so that the zirconia restoration and the dentin hard tissues can form good bond strength.

Declaration of competing interest

The authors have no conflicts of interest relevant to this article.

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