Journal of Prosthodontic Research

Official Journal of Japan Prosthodontic Society

Original article

Comparison of the shear bond strength of composite resins with zirconia and titanium using different resin cements

Pravej Serichetaphongse^{a,b,*}, Sirawit Chitsutheesiri^c, Wareeratn Chengrapakorn^{a,b}

^a Esthetics Restorative and Implant Dentistry Program, Faculty of Dentistry, Chulalongkorn University, Bangkok, Thailand

^b Department of Prosthodontics, Faculty of Dentistry, Chulalongkorn University, Bangkok, Thailand

^c Private practice, Dental hospital, Bangkok, Thailand

Abstract

Purpose: The shear bond strength of conventional zirconia (3Y-TZP), translucent zirconia (5Y-PSZ), and titanium alloy (Ti6Al4V) thermocycled using different phosphate monomer resin cements were investigated.

Methods: In this study, 120 specimens of 3Y-TZP, 5Y-PSZ, and Ti6Al4V were cemented to nanocomposite resin cylinders using PANAVIA™ V5 and Rely X™ U200. The bond area and resin cement thickness were controlled as per ISO 29022:2013 and 4049:2009. Each resin cement group was used with/without the Clearfil ceramic primer plus. The shear bond strength of the 12 groups was statistically analyzed using two and one-way ANOVA to determine the properties of the different materials and resin cements ($\alpha = 0.05$). The mode of failure was observed using scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS).

Results: The titanium alloy groups showed better shear bond strength than the zirconia groups ($p < 0.05$). PANAVIA™ V5 without primer showed significantly lower shear bond strength than other cements in zirconia and titanium alloy specimens ($p < 0.05$). Titanium alloy with Rely X™ U200 with a Clearfil ceramic primer plus showed the highest shear bond strength (6.37 ± 1.60 MPa). SEM images showed mixed failures in zirconia groups and cohesive failures in titanium alloy groups.

Conclusions: The titanium alloy showed better shear bond strength than zirconia when the Clearfil ceramic primer plus was used. The primer solution containing MDP and resin cement with phosphoric methacrylate ester showed similar shear bond strength with 3Y-TZP and 5Y-PSZ. The resin cement without phosphate monomers demonstrated the least shear bond strength.

Keywords: Zirconia, Titanium alloy, Implant abutment, Shear bond strength, Resin cement

Received 3 December 2020, Accepted 1 March 2021, Available online 25 June 2021

1. Introduction

Presently, zirconia is widely used in conventional restorations and implant dentistry [1]. Zirconia is used as monolithic crowns and as core materials layered with feldspathic porcelain to achieve the best esthetic outcome. Zirconia is a polymorphic, polycrystalline ceramic material with no glass components, resulting in superior properties compared with other glass ceramics [2]. Zirconia can be classified according to its composition and resulting translucency. Pure zirconia is in the monoclinic phase (M) at room temperature and undergoes a phase change to tetragonal phase (T) and a cubic phase (C) when fired at 1170 °C and 2370 °C, respectively. Stabilized oxides are added to help prevent the phase transformation during temperature changes, resulting in stabilized zirconia at room temperature. To stabilize tetragonal zirconia, 2–3 mol% yttrium oxide (Y_2O_3) is used which forms in 3 mol% yttria-partially stabilized zirconia (3Y-TZP) [3]. In contrast, highly translucent zirconia is stabilized by 4–8 mol% of yttrium oxide (Y_2O_3), leading to the formation of more cubic phases [4–9]. The

addition of 4–5 mol% of Y_2O_3 resulted in partially stabilized cubic phase zirconia, referred to in this study as 4 mol% yttria-partially stabilized zirconia (4Y-PSZ) and 5 mol% yttria-partially stabilized zirconia (5Y-PSZ). In addition 8 mol% Y_2O_3 [9]. Conventional 3Y-TZP has shown the highest flexural strength at 900–1,200 MPa [3] but lacks translucency [4]. Highly translucent zirconia has been developed to achieve better esthetics, but the flexural strength is reduced to 400–700 MPa [6,10,11]. Cubic zirconia has highly translucent characteristics owing to its isotropic crystallographic structure.

Titanium alloy (Ti₆Al₄V) is commonly used in implant dentistry because of its high strength-to-weight ratio, biocompatibility, and passivation. Titanium passivation is the formation of an oxide layer on the surface of the alloy when it is exposed to water or air. Passivation leads to high corrosion resistance in titanium [12].

Titanium-based (Ti-base) abutments with zirconia crowns are gaining popularity owing to the availability of Ti-base abutment configuration in design software, such as the 3shape dental system, and the Dental Wings open system. Furthermore, Ti-base abutments have a higher fracture resistance than zirconia abutments [13]. Titanium abutments have good mechanical properties; however, their use may compromise esthetics in patients with thin tissue layers. Consequently, titanium abutments are typically used with ceramic materials such as ceramic copings or crowns to improve esthetics [14]. The final restoration can be digitally designed using computer-aided design software, and the zirconia crown can be fabricated

* Corresponding author at: Esthetics Restorative and Implant Dentistry Program, Faculty of Dentistry, Chulalongkorn University, No. 34, Henry Dunant Road, Wangmai Pathumwan, Bangkok 10330, Thailand.

E-mail address: pravejs@hotmail.com (P. Serichetaphongse).

prior to purchasing the Ti-base abutment. Moreover, the Ti-base abutment could be used to rectify the angulation of dental implants up to 25° with a special screwdriver.

The bonding between zirconia and titanium material is another important factor for attaining the clinical success of Ti-base abutment cement-retained zirconia crowns. Typically, luting cement is bonded to the retainer and the abutment to seal the inevitable marginal gap. However, both titanium and zirconia materials have difficulty in bonding to all types of cement owing to their chemico-physical structure. Glass ceramics have shown the strongest bonding with resin cement due to the presence of the silica components in glass materials. Although zirconia is categorized as a ceramic material, it does not contain silica components. Therefore, hydrofluoric acid and silane coupling agents are ineffective [15,16]. Conventional cements such as zinc-phosphate cement, glass ionomer cement, resin-modified glass ionomer cement, and bisphenol A-glycidyl methacrylate (bis-GMA) resin cement have been reported to have weak bonding in zirconia [17,18]. Previous studies suggest that surface treatment of zirconia with Al_2O_3 particle air abrasion creates micromechanical retention. The chemical bonds in zirconium oxide on zirconia surfaces can be acquired by surface treatment along with phosphate-based functional monomers. This procedure should be performed to obtain durable and effective bonds [19,20] which are comparable with the bonding in metal alloys [21]. Various kinds of phosphate monomers have been included in resin cements and primers, including 10-methacryloyloxydecyl dihydrogen phosphate (MDP), phosphate methacrylate ester, glycerol phosphate dimethacrylate (GPDM), dipentaerythritol penta-acrylate monophosphate (PENTA), and phosphoric acid acrylate, all of which contribute to the enhancement of bond strength of zirconia [16,18,19,22–25]. Previous studies have reported shear bond strengths of zirconia to composite resins ranging from 2 to 25 MPa [20,24]. With the application of appropriate surface treatment and resin cements, 5Y-PSZ showed no differences in shear bond strength when compared with conventional 3Y-TZP [10].

In contrast, a study reported that polycarboxylate cement, zinc-phosphate cement, glass ionomer cement, and resin cement could provide effective bonding in pure titanium [26] and titanium alloy [27–29]. The use of Al_2O_3 particle air abrasion and phosphate monomer resin cement has been reported to provide improved bond strength to pure titanium [26] and titanium alloy [27–29]. Similar to zirconia, both micromechanical and chemical retention are required for strong bonding in titanium and titanium alloy. Studies reported so far demonstrate various shear bond strengths of titanium alloys in composite resins ranging from 12 to 25 MPa [26,30]. The luting cement for titanium must be chosen with caution, considering that few luting cements contain fluorides, such as polycarboxylate cement (Durelon and Poly-F plus) and glass ionomer cements (Fuji Slow Set, HY-Bond Glass Ionomer CX, Fuji Lute and Vitremer), which could lead to corrosion and discoloration of titanium oxide layer [27,31].

Since monolithic zirconia and titanium abutments have been more prevalent in esthetic and implant dentistry, reliable bonding of zirconia and titanium is essential for long-term clinical success. Therefore, the aim of this study was to investigate the shear bond strength of 3Y-TZP, 5Y-PSZ, and Ti6Al4V using various phosphate monomer resin cements by thermocycling. This study will be beneficial for the appropriate selection of resin cements for luting zirconia and titanium in dental restoration.

2. Materials and methods

2.1. Specimen preparation, fabrication, and characterization

A total of 120 cylindrical-shaped specimens of diameter 8.5 mm and height 3.2 mm were prepared from highly translucent zirconia (5Y-PSZ) (Ceramill® Zolid fx White; AMANN GIRRACH, Austria), slightly translucent zirconia (3Y-TZP) (Ceramill® Zi; AMANN GIRRACH, Austria), and titanium alloy (Ti6Al4V) (Trumpf®; TRUMPF GmbH+CO. KG., Germany). Both zirconia specimens (5Y-PSZ and 3Y-TZP) were milled from zirconia disks 30% larger than final cylindrical shape using a milling machine (Ceramill® Mikro 5, AMANN GIRRACH, Austria). The zirconia specimens were polished with silicon carbide (SiC) paper grit #1200 with a polishing machine (MINITECH 233, PRESI, France)

and then sintered using a sintering machine (Ceramill® Therm3, AMANN GIRRACH, Austria). On the other hand, titanium alloy cylindrical-shaped specimens were prepared using a laser-sintering machine (Truprint® 5000, TRUMPF GmbH+CO. KG, Germany) and polished using SiC paper grit #400, #800, and #1,200 in a polishing machine (Fig. 1).

All specimens were embedded in an autopolymerizing clear acrylic resin placed inside polyvinylchloride (PVC) tubes and polished with SiC paper grit #1200 in a polishing machine. Subsequently, the surface roughness of all the specimens was investigated using a surface roughness tester (Talyscan 150, England).

All samples were air-abraded for 15 s in a sandblasting unit (Basic Quattro, Renfert, Germany) using 50 μm Al_2O_3 particles at a pressure of 2.5 bar, with a perpendicular distance of 10 mm between the air abrasion instrument and the target surface [32]. Following this, the specimens were cleaned with 99% isopropanol in an ultrasonic bath (VGT-1990QTD, China) for 10 min.

To characterize the crystal phase content in 5Y-PSZ and 3Y-TZP, a sample from each zirconia specimen group was randomly selected and analyzed using an X-ray diffractometer (D8 Discover, Bruker AXS GmbH, Germany) with Cu-K α as the source at 40 kV and 40 mA. The surface of the specimens was analyzed within the range of 20° to 80° (2 θ) with a step size of 0.02° by an isolated observer (Fig. 2).

Moreover, 120 cylindrical-shaped nanocomposite resin samples Z350XT (Shade A3 BODY, 3M-ESPE, USA) with a diameter of 3.0 mm and a height of 2.0 mm were fabricated and light-cured using an LED light-curing unit (Demi Plus, Kerr, USA) with an intensity of 1,100 mW/cm² for 40 s.

2.2. Cementation procedure

All specimens were cemented under controlled bonding area and thickness as per ISO 4049:2009 [33] and ISO 29022:2013 [34]. An adhesive tape (thickness = 50 μm) with an inner circular hole (diameter = 2.38 mm) was placed on all specimens in order to obtain maximum resin cement thickness and bonding area.

Each specimen group was divided into four subgroups of ten depending on the cement type and primer inclusion as follows (Fig. 1): group 1 was composed of PANAVIA™ V5 (PV5; Dual-cured, Self-etching, Shade Universal (A2), Kuraray Noritake, Japan) with no primer, group 2 contained RelyX™ U200 (RXU; Dual-cured, Self-adhesive, Shade A2, 3M-ESPE, USA) with no primer; group 3 composed of PANAVIA™ V5 with Clearfil ceramic primer plus (PV5-C; Kuraray Noritake, Japan), and group 4 consisted of RelyX™ U200 with Clearfil ceramic primer plus (RXU-C). Subsequently, cementation of the specimens and the nanocomposite was performed according to the conditions stated for each group. The cylindrical-shaped nanocomposite resins were loaded with a 10 N weight using a durometer (ASTM D 2240 TYPE A, D PTC instruments, USA). The excess cement was removed with a microbrush and then light-cured with an LED light-curing unit (Demi Plus, Kerr, USA) with an intensity of 1,100 mW/cm² for 40 s (20 per side). All specimens were stored in distilled water at 37°C for 24 h and then subjected to thermocycling between 5°C and 55°C with a dwell time of 30 s and a transfer time of 5 s for 5,000 cycles [35]. The materials and resin cements used in this study are listed in Table 1.

2.3. Cementation procedure

After completion of thermocycling, all specimens were tested using a universal testing machine (EZ-S, SHIMADZU, Japan) with a notch-edge blade of the crosshead, 0.5 mm at the speed of 1 mm/min as per ISO 29022:2013 [34]. The shear bond strength R (MPa) was calculated from the expression $R=F/A$, where F is the load for specimen failure (N), and A is the cross-sectional interfacial area (mm²). Following the debonding of the specimens, the zirconia and titanium alloy surfaces were observed under a scanning electron microscope (SEM; FEI Quanta 250) at 2000× magnification to generate micrographs and to classify the mode of failure as adhesive, cohesive, or mixed failure. Adhesive failure occurs between the interfaces of the specimens and resin cement, cohesive failure occurs

Fig. 1. Experimental procedure schematic.

in the resin cement, and mixed failure is a combination of adhesive and cohesive failures. Energy-dispersive X-ray spectroscopy (EDS) was employed to identify the remnants on the zirconia and titanium alloy surfaces.

2.4. Statistical analysis

The data were statistically analyzed using IBM SPSS Statistics for Windows (version 22.0; IBM Corp, Armonk, NY, USA) statistics analysis program. Two-way ANOVA was used to analyze the effect of different types of material and resin cement on the shear bond strength of the specimens. One-way ANOVA and Tukey post hoc test or Welch and Games–Howell were used to determine the differences between the types of material and types of resin cement ($\alpha = 0.05$).

3. Results

The crystal phase contents of randomly selected high-translucent and low-translucent cylindrical zirconia specimens were evaluated using X-ray diffraction, as shown in Fig. 2, which depicts true 5Y-PSZ and 3Y-TZP zirconia specimens.

The shear bond strength test of the three materials (highly translucent zirconia [ZrHt], low translucent zirconia [ZrLt], and titanium alloy [TiA]) and four cementation procedures (PANAVIA™ V5 with and without Clearfil ceramic primer plus; and RelyX™ U200 with and without Clearfil ceramic primer plus) demonstrated that there were interactions between the various material and resin cement types according to two-way ANOVA, as presented in Table 2 ($p < 0.001$). The results of shear bond strength test are presented in Fig. 3. Titanium alloy presented the highest shear bond strength among the three materials when cemented with PANAVIA™ V5 and RelyX™ U200; both these cements contain Clearfil ceramic primer plus ($p < 0.05$). The TiA-RXU-C group showed the highest shear bond strength (6.37 ± 1.60 MPa).

Among the four different cementation procedures, the PV5 group showed the lowest shear bond strength compared to other resin cements for all material types ($p < 0.05$). In the zirconia materials group, no statistically significant variation could be identified between RXU, PV5-C, and RXU-C. However, statistically significant differences could be seen in resin cements in the TiA groups, where, RXU-C showed the highest shear bond strength and the PV5 group indicated the least.

The fractured surfaces were examined using SEM at 2,000× magnification. The mode of failure for each group is presented in Table 3. The ZrHt and ZrLt groups demonstrated adhesive and cohesive failures (mixed failure), while TiA groups showed predominantly cohesive failures in all groups of resin cement. The EDS results showed that silica (Si), sodium (Na), calcium (Ca), and barium (Ba) existed in the remnant islands on the zirconia surfaces and throughout the titanium surfaces (Fig. 4). SEM and EDS micrographs showed a higher mean-percentage of resin cement remnants ($48 \pm 13\%$ and $37 \pm 9\%$) in all RXU-C groups.

4. Discussion

To evaluate the shear bond strength in resin cement with zirconia and titanium alloy surfaces independently, the nanocomposite resin was utilized as a medium for the shear bond strength test. The results show that all cohesive failures were observed in the titanium alloy group (Table 3 and Fig. 4). Previous studies using a lower magnification (30–40×) optical microscope showed that the majority of adhesive failures occur in zirconia [24,36,37]. In contrast, a study conducted by Lee et al. [24] showed by observation of fractured surfaces using field-emission SEM analysis under 10,000× magnification that most samples predominantly underwent cohesive failures [24]. In this study, adhesive failures were observed at 50× magnification, however, at a magnification of 2,000×, mixed and cohesive failures were observed, as shown in Fig. 4.

The PANAVIA™ V5 cement, which does not contain a phosphate group, showed the lowest shear bond strength in all the materials studied. However, on the application of Clearfil ceramic primer plus, which contains MDP phosphate monomer, the shear bond strength of PANAVIA™ V5 increased in both zirconia and titanium alloys. This result agrees with those of previous studies that reported that the phosphate monomer in resin cement or primer improves the bond strength of zirconia and titanium alloys [24,38]. The use of phosphate monomer resin cement resulted in higher bond strength in both short- and long-term water storage. Moreover, it provided more durable bonds even after thermocycling [18,22]. Non-phosphate monomer resin cement yielded poor bond strength in 3Y-TZP [23], even when the surface was treated with air abrasion [16,39].

Al₂O₃ particle air abrasion with phosphate monomer resin cement or primer provides sustainable and durable bond strength on translucent zirconia [40,41], which is due to the chemical interaction between the P–OH group of the phosphate monomer and Zr–OH group on the zirconia surface [42]. Several previous studies also reported that resin cement or primers containing MDP could increase the bond strength by approximately 8 to 72 MPa on 3Y-TZP [16,18,19,22,23] and translucent zirconia [40,41]. Phosphoric methacrylate ester has also been reported to increase the bond strength of 3Y-TZP [24,25]. The results of this study showed that the shear bond strength of the primer containing MDP and the adhesive containing phosphoric methacrylate ester showed no significant differences, which was similar to the results of previous studies [37,43]. However, dissimilarly, MDP was reported to provide statistically higher shear bond strength than phosphoric methacrylate ester (4.62 ± 0.60 and 2.36 ± 0.41 MPa) [24], possibly due to the hydrolysis of methacrylate ester bonds in the presence of water [44].

In this study, 50 µm Al₂O₃ particles were used to air-abrade the specimens for 15 s at a pressure of 2.5 bar with a distance of 10 mm (perpendicular) between the air abrasion instrument and the target surface. This specificity of air abrasion covers approximately 32 mm² of the area. Therefore, a zirconia crown with an inner surface area of approximately 170 mm² might require at least 75 s to abrade the entire surface. However, a longer duration of abrasion could affect the roughness and phase transformation of zirconia [45].

Fig. 2. XRD patterns of zirconia specimens as interpreted by the International Centre for Diffraction Data database, demonstrating that highly translucent zirconia (ZrHt) contained zirconium oxide with cubic crystal phase. The single peaks (200, 220, 311 and 400)c are shown (A). Similarly, it was demonstrated that low translucent zirconia (ZrLt) contained zirconium oxide with tetragonal crystal phase. The doublet peaks (002, 200, 202, 220, 113, 311, 004 and 400)t are shown (B).

Table 1. Materials, abbreviations, manufacturers, compositions, and batch numbers.

Material	Abbreviation	Manufacturer	Composition	Batch No.
5Y-PSZ Ceramill® Zolid fx White	ZrHt	AMANN GIRRBACH AG, Austria	ZrO ₂ + Y ₂ O ₃ + HfO ₂ ≥99%wt, Y ₂ O ₃ 9.15 – 9.55%wt, HfO ₂ ≤5%wt, Al ₂ O ₃ ≤0.5%wt, Other oxides≤1%wt	1705001
3Y-TZP Ceramill® Zi	ZrLt	AMANN GIRRBACH AG, Austria	ZrO ₂ + Y ₂ O ₃ + HfO ₂ ≥99%wt, Y ₂ O ₃ 4.5 – 5.6%wt, HfO ₂ ≤5%wt, Al ₂ O ₃ ≤0.5%wt, Other oxides≤1%wt	1710001
Titanium alloy (Ti6Al4V ELI) Trumpf®; Laser sintering	TiA	TRUMPF GmbH+C0. KG., Germany	88-91% Ti, 5.5-6.5% Al, 3.5-4.5% V, <0.25% Fe, <0.13% O, <0.08% C, <0.05% N, <0.012% H	17-E361/15-45
Nanocomposite resin Filtek™ Z350 XT; Shade A3 BODY	-	3M ESPE, USA	Silane treated ceramic, Silane treated silica, UDMA, Bis-EMA, Bis-GMA, Silane treated zirconia, PEGDMA, TEGDMA	N991457
PANAVIA™ V5; Shade Universal (A2)	PV5	Kuraray Noritake, Japan	Paste: Bis-GMA, TEGDMA, Silanated barium glass filler, Silanated fluoroaluminosilicate glass filler, Colloidal silica, Surface treated aluminum oxide filler, Hydrophobic aromatic dimethacrylate, Hydrophilic aliphatic dimethacrylate, dl-Camphorquinone, Initiators, Accelerators, Pigments	5T0104
Clearfil ceramic primer plus	C	Kuraray Noritake, Japan	3-Methacryloxypropyl trimethoxysilane, MDP, Ethanol	8T0038
RelyX™ U200; Shade A2	RXU	3M ESPE, USA	Base: Glass powder, surface modified with 2-propenoic acid, 2 methyl-3-(trimethoxysilyl) propyl ester and phenyltrimethoxy silane bulk material, 2-propenoic acid, 2-methyl-, 1,1'-[1-(hydroxymethyl)-1,2-ethanediyl] ester reaction products with 2-hydroxy-1,3-propanediyl dimethacrylate and phosphorus oxide, TEGDMA, Silane treated silica, Oxide glass chemicals(nonfibrous), Sodium persulfate, Tert-butyl peroxy-3,5,5-trimethylhexanoate, Acetic acid, copper(2+) salt, monohydrate Catalyst: Glass powder, Surface modified with 2-propenoic acid, 2 methyl-3-(trimethoxysilyl) propyl ester, Bulk material, Substituted dimethacrylate, 1,12-dodecane dimethacrylate, Barbituric acid derivate, Silane treated silica, Sodium p-toluenesulfonate, Calcium hydroxide, Methacrylated aliphatic amine, 2,6-di-tert-butyl-p-cresol, 2-propenoic acid, 2-methyl-, 2-[(2-hydroxyethyl)(3-methoxypropyl)amino]ethyl ester, Titanium oxide	4819681

Table 2. Two-way ANOVA.

Source	Type III Sum of Squares	df	Mean Square	F	Sig.	Partial Eta Squared
Corrected Model	273.791 ^a	11	24.89	53.823	<0.001	0.846
Intercept	370.914	1	370.914	802.081	<0.001	0.881
material	77.509	2	38.755	83.805	<0.001	0.608
resin_cement	85.809	3	28.603	61.852	<0.001	0.632
material * resin_cement	110.473	6	18.412	39.815	<0.001	0.689
Error	49.943	108	0.462			
Total	694.649	120				
Corrected Total	323.735	119				

a. R Squared = .846 (Adjusted R Squared = .830)

Fig. 3. Mean and SD of shear bond strength of resin cements on highly translucent zirconia (ZrHt), low translucent zirconia (ZrLt) and titanium alloy (TiA). Different symbols indicate significant differences between resin cements and material types ($p < 0.05$).

Titanium alloys, similar to zirconia, also require Al_2O_3 particle air abrasion with phosphate monomer for stronger bonding, as a result, P-OH group were chemically bonded to titanium oxides [29,46]. A higher shear bond strength was observed with Rely XTM U200 incorporated with Clearfil ceramic primer plus on titanium alloy. Meanwhile, this primer showed no effect with Rely XTM U200 on zirconia, since no significant differences in the shear bond strength were observed in RXU and RXU-C on zirconia. This is because the amount of phosphoric methacrylate ester present in Rely XTM U200 was sufficient to form P-O-Zr bonds and to provide the needed bond strength in zirconia [43] due to the certain amount of Zr-OH on the zirconia surface. The formation of P-O-Zr between RXU and RXU-C might be similar. On the other hand, the titanium surface, which is rich in titanium oxide, and has a higher number of P-OH groups might increase the formation of P-O-Ti, resulting in improved bonding. Air abrasion might also increase wettability [26], which promotes improved bond strength. Therefore, air abrasion on titanium is recommended to achieve a better bond strength.

Both pure titanium and titanium alloys can be corroded under low pH by fluoride [12] unless the titanium alloy contains silver (Ag) or copper (Cu) [47]. Exposure to 0.1% sodium fluoride (NaF) or 452.4 ppm fluoride solution at pH 4.2, was hazardous to titanium alloy due to the formation of HF [48]. Polycarboxylate cement (Durelon and Poly-F plus) and glass ionomer cements (Fuji Slow Set, HY-Bond Glass Ionomer CX, Fuji Lute and Vitremer), which contain fluoride, can cause pitting corrosion and discolorations [27,31]. PANAVIATM V5 which is widely used for cementation of zirconia crowns, also contains fluoride, therefore, it

Table 3. Mode of failure at 2,000x magnification using SEM and mean percentage of resin cement remnant of each group.

Test groups	Mode of failure at 2,000x SEM			
	Adhesive (n)	Cohesive (n)	Mixed	
			(n)	Percentage of resin cement remnant, Mean ± SD (%)
ZrHt-PV5	3	0	7	21 ± 4
ZrHt-RXU	0	0	10	34 ± 7
ZrHt-PV5-C	0	2	8	48 ± 11
ZrHt-RXU-C	0	0	10	48 ± 13
ZrLt-PV5	1	0	9	13 ± 5
ZrLt-RXU	0	0	10	26 ± 10
ZrLt-PV5-C	0	3	7	33 ± 11
ZrLt-RXU-C	0	0	10	37 ± 9
TiA-PV5	0	10	0	-
TiA-RXU	0	10	0	-
TiA-PV5-C	0	10	0	-
TiA-RXU-C	0	10	0	-

Fig. 4. SEM images at 2,000x magnification and EDS of highly translucent zirconia (ZrHt), low translucent zirconia (ZrLt) and titanium alloy (TiA) with PANAVIATM V5 without and with Clearfil ceramic primer plus (PV5, PV5-C, respectively), and RelyXTM U200 without and with Clearfil ceramic primer plus (RXU, RXU-C, respectively). Predominantly mixed failure and cohesive failure were observed on zirconia and titanium alloy, respectively. Si, Na, Ca, and Ba existed in the remnant islands on zirconia surfaces and thoroughly on titanium surfaces. The white arrows indicate resin cement remnant on specimens with mixed failures.

should be used with caution or avoided. Other resin cements that contain phosphate monomers with no release of fluoride (e.g., the Rely X U200) should be used instead for cementation of Ti-base abutments with zirconia crowns.

The shear bond strength of composite resin on zirconia and titanium, which were 1–1.5 and 1–6 MPa, respectively, were very low compared to those of glass ceramics (10–20 MPa) [49,50]. Thus, the longevity of this type of restoration depends on how well the intaglio of the zirconia crown fits to the titanium abutment with sufficient resistance (abutment height > 5 mm). The controlled cement thickness of 50 μm as per ISO 4049:2009 resulted in lower shear bond strength of zirconia and titanium compared to those reported in earlier studies [33]. Moreover, the thermocycling procedure could weaken the resin cement via the hydrolysis effect, resulting in cohesive failures [20] due to the bonding degeneration between

the filler and matrix in resin cements [51]. Nonetheless, thermocycling is an appropriate procedure for simulating the intraoral aging of resin cements via repeated cyclic exposure to temperature changes in an aqueous environment [52].

According to the EDS analysis of the fractured specimen, remnants of Si, Na, Ca, and Ba existed on the surfaces of the zirconia and titanium specimens. These existing elements were the components of the resin cement used in each group, as shown in Table 1. This analysis confirmed the mode of failure for each specimen groups. In the PV5 group, resin cement remnants were present on the specimens, which might have resulted from micromechanical retention created by air abrasion. In the Clearfil ceramic primer plus groups, the bond capacity of resin cements was improved due to chemical bonding of the P-OH group and improvement in the wettability of the primer after air abrasion [53].

The limitations of this study are as follows: firstly, only limited types of resin cements were used and secondly, the Clearfil primer used in this study was not a pure MDP solution but contained silane and MDP. A pure MDP solution should be used in future work. Finally, the 5,000-thermocycling cycles considered in this study were not sufficient to predict the longevity of the bonding.

5. Conclusion

Within the limitations of this study, the following conclusions can be drawn. The addition of the phosphate monomer improved the shear bond strength of both titanium and zirconia. Titanium alloy demonstrated better shear bond strength to resin cements compared to zirconia when the Clearfil ceramic primer plus (MDP-containing primer) was applied. Non-phosphate monomer resin cement (PANAVIA™ V5) must be used along with the Clearfil ceramic primer plus to bond with zirconia and titanium. Moreover, the type of zirconia had no influence on the shear bond strength when resin cement is used. Therefore, when zirconia is cemented with a titanium alloy along with a phosphate monomer resin cement, the Clearfil ceramic primer plus should be applied on the titanium side.

Acknowledgements

The authors would like to express their gratitude to the Oral Biology Research and Dental Materials R&D Center, Faculty of Dentistry, Chulalongkorn University, for allowing the use of their facility. Dr. Naruedee Limpuangthip Department of Prosthodontics, Faculty of Dentistry, Chulalongkorn University, kindly helped with the statistical analysis. This study did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Conflict of interest

The authors declare no competing financial interests or personal relationships that could have appeared to influence the results reported in this paper.

References

- [1] Kelly JR, Benetti P. Ceramic materials in dentistry: Historical evolution and current practice. *Aust Dent J* 2011;56:84-96. <https://doi.org/10.1111/j.1834-7819.2010.01299.x>
- [2] Kelly JR, Denry I. Stabilized zirconia as a structural ceramic: An overview. *Dent Mater* 2008;24:289-98. <https://doi.org/10.1016/j.dental.2007.05.005>
- [3] Piconi C, Maccauro G. Zirconia as a ceramic biomaterial. *Biomaterials* 1999;20:1-25. [https://doi.org/10.1016/S0142-9612\(98\)00010-6](https://doi.org/10.1016/S0142-9612(98)00010-6)
- [4] Zhang Y. Making yttria-stabilized tetragonal zirconia translucent. *Dent Mater* 2014;30:1195-203. <https://doi.org/10.1016/j.dental.2014.08.375>
- [5] Zhang F, Inokoshi M, Batuk M, Hadermann J, Naert I, Van Meerbeek B, et al. Strength, toughness and aging stability of highly-translucent Y-TZP ceramics for dental restorations. *Dent Mater* 2016;32:e327-e37. <https://doi.org/10.1016/j.dental.2016.09.025>
- [6] Pereira GKR, Guillard LF, Dapieve KS, Kleverlaan CJ, Rippe MP, Valandro LF. Mechanical reliability, fatigue strength and survival analysis of new polycrystalline translucent zirconia ceramics for monolithic restorations. *J Mech Behav Biomed Mater* 2018;85:57-65. <https://doi.org/10.1016/j.jmbbm.2018.05.029>
- [7] Inokoshi M, Shimizu H, Nozaki K, Takagaki T, Yoshihara K, Nagaoka N, et al. Crystallographic and morphological analysis of sandblasted highly translucent dental zirconia. *Dent Mater* 2018;34:508-18. <https://doi.org/10.1016/j.dental.2017.12.008>
- [8] Lange F. Transformation Toughening: Part 3, Experimental Observations in the ZrO₂-Y₂O₃ System. *J Mater Sci* 1982;17:240-6. <https://doi.org/10.1007/BF00809059>
- [9] Ghodsi S, Jafarian Z. A Review on Translucent Zirconia. *Eur J Prosthodont Restor Dent* 2018;26:62-74. https://doi.org/10.1922/EJPRD_01759Ghodsi13
- [10] Kwon SJ, Lawson NC, McLaren EE, Nejat AH, Burgess JO. Comparison of the mechanical properties of translucent zirconia and lithium disilicate. *J Prosthet Dent* 2018;120:132-7. <https://doi.org/10.1016/j.prosdent.2017.08.004>
- [11] Nassary Zadeh P, Lumkemann N, Sener B, Eichberger M, Stawarczyk B. Flexural strength, fracture toughness, and translucency of cubic/tetragonal zirconia materials. *J Prosthet Dent* 2018;120:948-54. <https://doi.org/10.1016/j.prosdent.2017.12.021>
- [12] Prasad S, Ehrensberger M, Gibson MP, Kim H, Monaco EA. Biomaterial properties of titanium in dentistry. *J Oral Biosci* 2015;57:192-9. <https://doi.org/10.1016/j.job.2015.08.001>
- [13] Foong JK, Judge RB, Palamara JE, Swain MV. Fracture resistance of titanium and zirconia abutments: an in vitro study. *J Prosthet Dent* 2013;109:304-12. [https://doi.org/10.1016/S0022-3913\(13\)60306-6](https://doi.org/10.1016/S0022-3913(13)60306-6)
- [14] Kim JS, Raigrodski AJ, Flinn BD, Rubenstein JE, Chung K-H, Mancel LA. In vitro assessment of three types of zirconia implant abutments under static load. *J Prosthet Dent* 2013;109:255-63. [https://doi.org/10.1016/S0022-3913\(13\)60054-2](https://doi.org/10.1016/S0022-3913(13)60054-2)
- [15] Matinlinna JP, Heikkinen T, Özcan M, Lassila LVJ, Vallittu PK. Evaluation of resin adhesion to zirconia ceramic using some organosilanes. *Dent Mater* 2006;22:824-31. <https://doi.org/10.1016/j.dental.2005.11.035>
- [16] Kern M, Wegner SM. Bonding to zirconia ceramic: adhesion methods and their durability. *Dent Mater* 1998;14:64-71. [https://doi.org/10.1016/S0109-5641\(98\)00011-6](https://doi.org/10.1016/S0109-5641(98)00011-6)
- [17] Piwowarczyk A, Lauer HC, Sorensen JA. The shear bond strength between luting cements and zirconia ceramics after two pre-treatments. *Oper Dent* 2005;30:382-8.
- [18] Lüthy H, Loeffel O, Hammerle CHF. Effect of thermocycling on bond strength of luting cements to zirconia ceramic. *Dent Mater* 2006;22:195-200. <https://doi.org/10.1016/j.dental.2005.04.016>
- [19] Wolfart M, Lehmann F, Wolfart S, Kern M. Durability of the resin bond strength to zirconia ceramic after using different surface conditioning methods. *Dent Mater* 2007;23:45-50. <https://doi.org/10.1016/j.dental.2005.11.040>
- [20] Grasel R, Santos MJ, Rego HC, Rippe MP, Valandro LF. Effect of Resin Luting Systems and Alumina Particle Air Abrasion on Bond Strength to Zirconia. *Oper Dent* 2018;43:282-90. <https://doi.org/10.2341/15-352-1>
- [21] Yoshida K, Kamada K, Tanagawa M, Atsuta M. Shear bond strengths of three resin cements used with three adhesive primers for metal. *J Prosthet Dent* 1996;75:254-61. [https://doi.org/10.1016/S0022-3913\(96\)90481-3](https://doi.org/10.1016/S0022-3913(96)90481-3)
- [22] Yoshida K, Tsuo Y, Atsuta M. Bonding of dual-cured resin cement to zirconia ceramic using phosphate acid ester monomer and zirconate coupler. *J Biomed Mater Res B Appl Biomater* 2006;77B:28-33. <https://doi.org/doi:10.1002/jbm.b.30424>
- [23] Llerena-Icochea AE, Costa RM, Borges A, Bombonatti J, Furuse AY. Bonding Polycrystalline Zirconia With 10-MDP-containing Adhesives. *Oper Dent* 2017;42:335-41. <https://doi.org/10.2341/16-156-1>
- [24] Lee S-E, Bae J-H, Choi J-W, Jeon Y-C, Jeong C-M, Yoon M-J, et al. Comparative Shear-Bond Strength of Six Dental Self-Adhesive Resin Cements to Zirconia. *Materials* 2015;8:3306-15. <https://doi.org/10.3390/ma8063306>
- [25] Liu W, Meng H, Sun Z, Jiang R, Dong C-A, Zhang C. Phosphoric and carboxylic methacrylate esters as bonding agents in self-adhesive resin cements. *Exp Ther Med* 2018;15:4531-7. <https://doi.org/10.3892/etm.2018.5937>
- [26] Abi-Rached Fde O, Fonseca RG, Haneda IG, de Almeida-Júnior AA, Adabo GL. The effect of different surface treatments on the shear bond strength of luting cements to titanium. *J Prosthet Dent* 2012;108:370-6. [https://doi.org/10.1016/S0022-3913\(12\)60194-2](https://doi.org/10.1016/S0022-3913(12)60194-2)
- [27] Wadhvani C, Chung K-H. Bond strength and interactions of machined titanium-based alloy with dental cements. *J Prosthet Dent* 2015;114:660-5. <https://doi.org/10.1016/j.prosdent.2015.04.015>
- [28] Mehl C, Zhang Q, Lehmann F, Kern M. Retention of zirconia on titanium in two-piece abutments with self-adhesive resin cements. *J Prosthet Dent* 2018;120:214-9. <https://doi.org/10.1016/j.prosdent.2017.11.020>
- [29] Kunt GE, Ceylan G, Yilmaz N. Effect of surface treatments on implant crown retention. *J Dent Sci* 2010;5:131-5. [https://doi.org/10.1016/S1991-7902\(10\)60019-0](https://doi.org/10.1016/S1991-7902(10)60019-0)
- [30] Ohkubo C, Watanabe I, Hosoi T, Okabe T. Shear bond strengths of polymethyl methacrylate to cast titanium and cobalt-chromium frameworks using five metal primers. *J Prosthet Dent* 2000;83:50-7. [https://doi.org/10.1016/S0022-3913\(00\)70088-6](https://doi.org/10.1016/S0022-3913(00)70088-6)
- [31] Horasawa N, Marek M. Effect of fluoride from glass ionomer on discoloration and corrosion of titanium. *Acta Biomater* 2010;6:662-6. <https://doi.org/10.1016/j.actbio.2009.08.019>
- [32] Hallmann L, Ulmer P, Reusser E, Hammerle CHF. Effect of blasting pressure, abrasive particle size and grade on phase transformation and morphological change of dental zirconia surface. *Surf Coat Technol* 2012;206:4293-302. <https://doi.org/10.1016/j.surfcoat.2012.04.043>
- [33] International Standards Organization. Technical specification 4049. Dentistry-Polymer-based restorative materials. Switzerland: International Standards Organization; 2009. ed., p. of.

- [34] International Standards Organization. Technical specification 29022. Dentistry-Adhesion-Notched-edge shear bond strength test. Switzerland: International Standards Organization; 2013. ed., p. of.
- [35] Ozcan M, Bernasconi M. Adhesion to zirconia used for dental restorations: a systematic review and meta-analysis. *J Adhes Dent* 2015;17:7-26. <https://doi.org/10.3290/j.jad.a33525>
- [36] Da Silva EM, Miragaya L, Sabrosa CE, Maia LC. Stability of the bond between two resin cements and an yttria-stabilized zirconia ceramic after six months of aging in water. *J Prosthet Dent* 2014;112:568-75. <https://doi.org/10.1016/j.prosdent.2013.12.003>
- [37] Mirmohammadi H, Aboushelib MN, Salameh Z, Feilzer AJ, Kleverlaan CJ. Innovations in bonding to zirconia based ceramics: Part III. Phosphate monomer resin cements. *Dent Mater* 2010;26:786-92. <https://doi.org/10.1016/j.dental.2010.04.003>
- [38] Kern M, Van Thompson P. Durability of Resin Bonds to Pure Titanium. *J Prosthodont* 1995;4:16-22. <https://doi.org/10.1111/j.1532-849X.1995.tb00309.x>
- [39] Kern M, Barloi A, Yang B. Surface Conditioning Influences Zirconia Ceramic Bonding. *J Dent Res* 2009;88:817-22. <https://doi.org/10.1177/0022034509340881>
- [40] Le M, Larsson C, Papia E. Bond strength between MDP-based cement and translucent zirconia. *Dent Mater J* 2019;38:480-9. <https://doi.org/10.4012/dmj.2018-194>
- [41] Yagawa S, Komine F, Fushiki R, Kubochi K, Kimura F, Matsumura H. Effect of priming agents on shear bond strengths of resin-based luting agents to a translucent zirconia material. *J Prosthodont Res* 2018;62:204-9. <https://doi.org/10.1016/j.jpor.2017.08.011>
- [42] Nagaoka N, Yoshihara K, Feitosa VP, Tamada Y, Irie M, Yoshida Y, et al. Chemical interaction mechanism of 10-MDP with zirconia. *Sci Rep* 2017;7:45563. <https://doi.org/10.1038/srep45563>
- [43] Passia N, Mitsias M, Lehmann F, Kern M. Bond strength of a new generation of universal bonding systems to zirconia ceramic. *J Mech Behav Biomed Mater* 2016;62:268-74. <https://doi.org/10.1016/j.jmbbm.2016.04.045>
- [44] Yoshihara K, Nagaoka N, Okihara T, Kuroboshi M, Hayakawa S, Maruo Y, et al. Functional monomer impurity affects adhesive performance. *Dent Mater* 2015;31:1493-501. <https://doi.org/10.1016/j.dental.2015.09.019>
- [45] Turp V, Sen D, Tuncelli B, Goller G, Özcan M. Evaluation of air-particle abrasion of Y-TZP with different particles using microstructural analysis. *Aust Dent J* 2013;58:183-91. <https://doi.org/10.1111/adj.12065>
- [46] Yanagida H, Matsumura H, Taira Y, Atsuta M, Shimoe S. Adhesive bonding of composite material to cast titanium with varying surface preparations. *J Oral Rehabil* 2002;29:121-6. <https://doi.org/10.1046/j.1365-2842.2002.00811.x>
- [47] Al-Mayouf AM, Al-Swayih AA, Al-Mobarak NA, Al-Jabab AS. Corrosion behavior of a new titanium alloy for dental implant applications in fluoride media. *Mater Chem Phys* 2004;86:320-9. <https://doi.org/10.1016/j.matchemphys.2004.03.019>
- [48] Nakagawa M, Matsuya S, Shiraishi T, Ohta M. Effect of fluoride concentration and pH on corrosion behavior of titanium for dental use. *J Dent Res* 1999;78:1568-72. <https://doi.org/10.1177/00220345990780091201>
- [49] Taguchi S, Komine F, Kubochi K, Fushiki R, Kimura F, Matsumura H. Effect of a silane and phosphate functional monomer on shear bond strength of a resin-based luting agent to lithium disilicate ceramic and quartz materials. *J Oral Sci* 2018;60:360-6. <https://doi.org/10.2334/josnusd.17-0383>
- [50] Lanza MDS, Rodrigues Lanza FJS, Manso AP, Matinlinna JP, Carvalho RM. Innovative surface treatments for improved ceramic bonding: Lithium disilicate glass ceramic. *Int J Adhes Adhes* 2018;82:60-6. <https://doi.org/10.1016/j.jadhadh.2017.12.007>
- [51] Soderholm KJ, Roberts MJ. Influence of water exposure on the tensile strength of composites. *J Dent Res* 1990;69:1812-6. <https://doi.org/10.1177/00220345900690120501>
- [52] Morresi AL, D'Amario M, Capogreco M, Gatto R, Marzo G, D'Arcangelo C, et al. Thermal cycling for restorative materials: does a standardized protocol exist in laboratory testing? A literature review. *J Mech Behav Biomed Mater* 2014;29:295-308. <https://doi.org/10.1016/j.jmbbm.2013.09.013>
- [53] Pereira Lde L, Campos F, Dal Piva AM, Gondim LD, Souza RO, Özcan M. Can application of universal primers alone be a substitute for airborne-particle abrasion to improve adhesion of resin cement to zirconia? *J Adhes Dent* 2015;17:169-74. <https://doi.org/10.3290/j.jad.a33974>

Copyright: This is an open-access article distributed under the terms of Creative Commons Attribution License 4.0 (CCBY 4.0), which allows users to distribute and copy the material in any format so long as attribution is given to the author(s).