

Original article

Effects of two methyl methacrylate-tributylborane-based luting agents with a silane-phosphate primer on bonding of four different CAD/CAM resin composite materials

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(Received November 27, 2021; Accepted February 24, 2022)

Abstract

Purpose: This study compared the bond strengths of four adhesive systems and four different resin composite block materials: Gradia Block (GR), Shofu Block HC (SH), Estelite Block (ES), and KZR-CAD HR2 (KZ).

Methods: A primer (PZ-AB) containing a silane (γ -MPTS) with 10-methacryloyloxydecyl dihydrogen phosphate (MDP) was applied to ground surfaces of the resin composite block specimens, and the specimens were then bonded to stainless-steel rods using two methyl methacrylate-tributylborane (MMA-TBB)-based luting agents (SB and MT), designated as the PZ-AB/SB and PZ-AB/MT adhesive systems, respectively. The SB resin contained 4-methacryloyloxyethyl trimellitate anhydride (4-META), whereas the MT resin did not. The SB resin without primer (No primer/SB) and a dual-curing composite-type adhesive system (UPA/RelyX) were used as controls. The 24-h tensile bond strengths were determined and analyzed using the Tukey-Kramer HSD test ($\alpha = 0.05$, $n = 8$).

Results: The highest bond strengths were obtained for the GR/PZ-AB/MT, GR/PZ-AB/SB, KZ/PZ-AB/MT, ES/PZ-AB/SB, and KZ/No primer/SB groups, whereas the KZ/UPA/RelyX, ES/UPA/RelyX, SH/UPA/RelyX, and SH/No primer/SB groups exhibited the lowest bond strengths.

Conclusion: For each resin composite block material primed with γ -MPTS and MDP, the MMA-TBB-based luting agents, irrespective of the presence of 4-META, provided higher bond strengths than the dual-curing composite-type adhesive system.

Keywords: bond strength, CAD/CAM, luting material, resin composite block

Introduction

The widespread use of computer-aided design and computer-aided manufacturing (CAD/CAM) systems in dentistry has resulted in increased opportunities to apply resin composites to indirect restorations such as crowns, inlays, and veneers [1-3]. Highly filled resin composite blocks are generally fabricated by infiltrating resin monomers in compressed filler particles under high pressure followed by heat-initiated polymerization. As reported previously, the mechanical properties of the resulting materials are acceptable for the fabrication of single restorations [4]. Compared to machinable ceramics or metal alloys, highly-filled resin composites are advantageous because they have comparable wear resistance to enamel, low potential to abrade the opposing tooth structure, and good machinability [4,5]. However, resin composite restorations that are bonded on an abutment tooth are occasionally detached or partly fractured [6,7], particularly when conservative preparation designs with minimal mechanical retentions are selected. Although multiple factors are responsible for such failures, the strength of adhesive bonding between the resin composite and the luting agent is an important factor that determines the ability of restorations to withstand the oral environment.

Surface treatments involving air-abrasion [8-11], silica coating [12], hydrofluoric acid etching and silanization [13], ceramic repair systems [14], and priming agents [15,16] have been reported to improve the bonding of resin-based materials to resin composite block materials. In a previous study, the application of an unfilled resin composed of methyl methacrylate (MMA) and a tributylborane (TBB) derivative in conjunction with silane improved the bonding to a resin composite block material [17,18]. Several commercially available composite-type luting agents also employ a priming agent containing silane. However, it remains unclear whether MMA-TBB resin or a composite-type luting agent bonds more strongly to the resin composite block material.

When bonding resin composite block materials with a self-curing luting agent (4-META/MMA-TBB resin) composed of 4-methacryloyloxyethyl trimellitate anhydride (4-META), MMA, and TBB derivative, a two-liquid primer (Super-Bond PZ primer, Sun Medical Co., Ltd., Moriyama, Japan) comprising a silane (3-trimethoxysilylpropyl methacrylate, γ -MPTS) and a phosphate monomer (10-methacryloyloxydecyl dihydrogen phosphate, MDP) is recommended by the manufacturer [19]. However, the efficacy of bonding between a silane-phosphate primer and resin composite block materials of varying chemical composition has yet to be investigated under identical conditions. Therefore, the purpose of this study was to determine the bond strengths between two MMA-TBB-based luting agents with and without 4-META and four different resin composite block materials primed with γ -MPTS and MDP, comparing the results with those obtained using a dual-curing composite-type adhesive system. The null hypothesis was that the bond strength would not be affected by either the type of adhesive system or the resin composite block material.

Materials and Methods

Materials

The substrate materials, priming agents, and luting agents used are summarized in Table 1. Gradia Block (GR; GC Corp., Tokyo, Japan), Shofu Block HC (SH; Shofu Inc., Kyoto, Japan), Estelite Block (ES; Tokuyama Dental Corp., Tokyo, Japan), and KZR-CAD HR2 (KZ; Yamakin Co., Ltd., Osaka, Japan) were employed as the substrate materials [4,18,20-22]. A two-bottle priming agent (Super-Bond PZ Primer, PZ-AB, Sun Medical Co., Ltd.) was used in conjunction with a 4-META/MMA-TBB resin (Super-Bond C&B, SB, Sun Medical Co., Ltd.) or an MMA-TBB resin (MT) without 4-META.

Preparation of the bonded specimens

A total of 128 rectangular specimens (32 specimens \times 4 resin composite block materials) with dimensions of 7 \times 12 \times 3 mm were cut from the resin composite blocks using a diamond saw (IsoMet Low Speed Saw, Buehler, Lake Bluff, IL, USA). All specimens were ground with 600-grit silicon-carbide abrasive paper (BuehlerMet2, Buehler), cleaned ultrasonically in a distilled water bath for 5 min, and air-dried. A piece of masking tape with a circular hole (diameter: 2 mm) was attached to the surface of each specimen to delineate the bonding area and the thickness of the luting agent (Fig. 1). The specimens for each resin composite block material were divided into four adhesive systems (No primer/SB, PZ-AB/SB, PZ-AB/MT, and UPA/RelyX) with eight for specimens each. The priming agents were applied to the specimens in the corresponding groups with an applicator brush and gently air-dried. The flat side of a stainless-steel rod (diameter: 5 mm, length: 20 mm) was air-abraded with alumina (HI-

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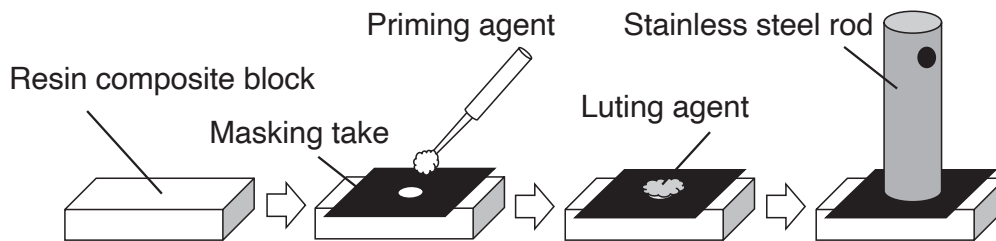
doi.org/10.2334/josnusd.21-0513

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Table 1 Substrate materials, priming agents, and luting agents used

Name (abbreviation), Color	Component	Filler* (mass%)	Manufacturer	Lot no.
Substrate material				
Gradia Block (GR), A3	UDMA 20%; multifunctional methacrylate 4%; organic-inorganic composite filler 3%; silica-based powder 73%	76	GC Corp., Tokyo, Japan	1410011
Shofu Block HC (SH), A3	UDMA; TEGDMA; silica; micro fumed silica; zirconium silicate	61	Shofu Inc., Kyoto, Japan	091501
Estelite Block (ES), A3	UDMA; TEGDMA; silica; zirconia	75	Tokuyama Dental Corp., Tokyo, Japan	006085
KZR-CAD HR2 (KZ), A3	UDMA; TEGDMA; aggregated SiO ₂ -ZrO ₂ -Al ₂ O ₃ (200-600 nm) cluster (1-20 μm); SiO ₂ (20 nm); fluoride filler (700 nm)	72	Yamakin Co., Ltd., Osaka, Japan	01101506
Priming agent				
Super-Bond PZ Primer (PZ-AB)	liquid-A: MDP; MMA liquid-B: γ-MPTS; MMA		Sun Medical Co., Ltd. Moriyama, Japan	TM1 TM12
Universal Plus Adhesive (UPA)	2-propenoic acid, 2-methyl-, diesters with 4,6-dibromo-1,3-benzenediol 2-(2-hydroxyethoxy) ethyl 3-hydroxypropyl diethers; HEMA; 2-propenoic acid, 2-methyl-, reaction products with 1,10-decanediol and phosphorus oxide; MDP; 2-propenoic acid, 2-methyl-, 3-(triethoxysilyl)propyl ester and (3-aminopropyl)triethoxysilane, reaction products with vitreous silica; ethanol; water; camphorquinone; copolymer of acrylic and itaconic acid; <i>N,N</i> -dimethylbenzocaine; (3-aminopropyl) triethoxysilane; acetic acid, copper salt, monohydrate		3M Deutschland GmbH, Neuss, Germany	7444306
Luting agent				
4-META/MMA-TBB resin (SB)	catalyst V: TBB derivative polymer powder opaque ivory: PMMA, titanium oxide monomer liquid: MMA, 4-META		Sun Medical Co., Ltd.	TF1 TF1 TG1
MMA-TBB resin (MT)	catalyst V: TBB derivative polymer powder opaque ivory: PMMA, titanium oxide monomer liquid: MMA		Sun Medical Co., Ltd. Fujifilm Wako Pure Chemical Corp., Osaka, Japan	TF1 TF1 APG0220
RelyX Universal Resin Cement (RelyX)	catalyst paste: UDMA; ytterbium (III) fluoride; glass powder; TEGDMA; L-ascorbic acid, 6-hexadecanoate, hydrate; silane, trimethoxyoctyl-, hydrolysis products with silica; HEMA; titanium dioxide; triphenyl phosphite base paste: γ-MPTS; UDMA; TEGDMA; mixture of mono- di- and tri-glycerol dimethacrylate ester of phosphoric acid; silane, trimethoxyoctyl-, hydrolysis products with silica; t-amyl hydroperoxide; 2,6-di-tert-butyl-p-cresol; HEMA; MMA; acetic acid, copper salt, monohydrate		3M Deutschland GmbH	7432332

UDMA, urethane dimethacrylate; TEGDMA, triethyleneglycol dimethacrylate; MDP, 10-methacryloyloxydecyl dihydrogen phosphate; MMA, methyl methacrylate; γ-MPTS, 3-trimethoxysilylpropyl methacrylate; HEMA, 2-hydroxyethyl methacrylate; TBB, tributylborane; PMMA, poly(methyl methacrylate); 4-META, 4-methacryloyloxyethyl trimellitate anhydride. *Ref.: Lauvahutanon et al. [4], Shinohara et al. [18], Kamonkhanitkul et al. [20], Asano et al. [21], Iwata et al. [22]

**Fig. 1** Schematic representation of the bonding procedure

Alumina, Shofu Inc.), primed with one component of the PZ-AB primer (liquid-A), and bonded to the resin composite specimen with the luting agent (MT or SB).

In addition to the SB resin without primer (No primer/SB), a commercially available dual-curing adhesive system (UPA/RelyX) composed of a priming agent (Universal Plus Adhesive, 3M Deutschland GmbH, Neuss, Germany) and a composite-type luting agent (RelyX Universal Resin Cement, 3M Deutschland GmbH) was used as a control in accordance with the manufacturer's instructions. The UPA/RelyX material was light cured with a light-emitting diode unit (Pencure, J Morita Corp., Tokyo, Japan) from both lateral sides of the bonded specimen for 10 s each.

Tensile bond strength tests

At 30 min after preparation, the bonded specimens were immersed in water at 37°C for 24 h. The tensile bond strength of each specimen was determined using a universal testing machine (AGS-10kNG, Shimadzu Corp., Kyoto, Japan) at a cross-head speed of 1.0 mm/min.

Failure mode observations

Following the tensile bond strength tests, the debonded surfaces of all specimens were observed with an optical microscope (SMZ-10, Nikon

Corp., Tokyo, Japan) at a magnification of ×20 to assess the bond failure. The failure modes were categorized as follows: Adhesive failure at the interface between the resin composite block material and the luting agent (Ad), cohesive failure within the luting agent (Co), or mixed failure comprising both the Ad and Co modes (Ad/Co).

Scanning electron microscopy (SEM) observations

The surfaces of the additional composite blocks were ground with 600-grit silicon-carbide abrasive paper, cleaned ultrasonically in a distilled water bath, air-dried, sputter-coated with gold (Ion Coater IB-3, Eiko Engineering Co., Ltd., Hitachinaka, Japan), and observed using a scanning electron microscope (JCM-6000Plus, JEOL Ltd., Tokyo, Japan) at a magnification of ×8,000. In addition, a debonded SH/PZ-AB/SB specimen was sputter-coated and observed using SEM at ×2,000 magnification.

Statistical analysis

The mean bond strengths and standard deviations were calculated for all sixteen test groups containing eight specimens each. The reliability of the sample size, normality, and assumption of homoscedasticity were verified using power analysis, the Anderson-Darling test, and the Brown-Forsythe test, respectively. The data were analyzed using the *post hoc* Tukey-Kramer

Table 2 Tensile bond strength between four adhesive systems and four resin composite block materials

Adhesive system		Bond strength (SD) (MPa)*			
Priming agent	Luting agent	GR	SH	ES	KZ
No primer	SB	16.8 (4.3) ^{bcd}	4.2 (2.1) ^e	15.0 (4.4) ^{def}	21.3 (4.1) ^{abc}
PZ-AB	SB	24.4 (6.8) ^{ab}	13.4 (3.2) ^{def}	21.9 (6.9) ^{abc}	20.1 (4.8) ^{bcd}
PZ-AB	MT	28.8 (5.5) ^a	13.2 (2.5) ^{def}	20.6 (2.4) ^{bcd}	24.1 (6.9) ^{ab}
UPA	RelyX	17.1 (4.3) ^{bcd}	4.9 (3.9) ^e	9.0 (2.8) ^{fg}	10.1 (2.0) ^{efg}

*Identical small letters indicate values that are not significantly different ($P \geq 0.05$).

Table 3 Two-way ANOVA results corresponding to Table 2

Factor	d.f.	Sum of squares	Mean square	F-value	P-value
Adhesive system	3	2631.0	877.0	43.9	<0.0001
Resin composite block material	3	2908.1	969.3	48.5	<0.0001
Adhesive system/Resin composite block material	9	532.9	59.2	3.0	0.003
Residual	112	2236.8	20.0		

Table 4 Failure modes observed after tensile bond testing

Adhesive system		Failure mode* (number of specimens)			
Priming agent	Luting agent	GR	SH	ES	KZ
No primer	SB	Ad/Co (8)	Ad/Co (8)	Ad/Co (8)	Ad/Co (8)
PZ-AB	SB	Ad/Co (7), Co (1)	Ad/Co (8)	Ad/Co (8)	Ad/Co (7), Co (1)
PZ-AB	MT	Ad/Co (5), Co (3)	Ad/Co (8)	Ad/Co (8)	Ad/Co (8)
UPA	RelyX	Ad (8)	Ad (8)	Ad (8)	Ad (8)

*Ad, adhesive failure at the interface between the resin composite block material and the luting agent. Co, cohesive failure within the luting agent. Ad/Co, mixed failure in the Ad and Co modes

HSD test following two-way ANOVA, with the threshold of statistical significance set at 0.05. The statistical analysis was carried out using the JMP Pro software system (Ver. 15, SAS Institute Japan Ltd., Tokyo, Japan).

Results

Tensile bond strength

The mean tensile bond strength ranged from 4.2 MPa (SH/No primer/SB) to 28.8 MPa (GR/PZ-AB/MT) (Table 2). The experimental groups GR/PZ-AB/MT, GR/PZ-AB/SB, KZ/PZ-AB/MT, ES/PZ-AB/SB, and KZ/No primer/SB had the highest bond strengths, whereas the KZ/UPA/RelyX, ES/UPA/RelyX, SH/UPA/RelyX, and SH/No primer/SB groups had the lowest. No significant difference was observed in the bond strengths of KZ/No primer/SB and KZ/PZ-AB/SB. For the three resin composite block materials, namely, GR, SH, and ES, the differences were not always significant; however, the PZ-AB/SB group tended to exhibit higher bond strength than the No primer/SB control. For the three resin composite block materials – SH, ES, and KZ – the PZ-AB/MT and PZ-AB/SB groups exhibited significantly higher bond strengths than UPA/RelyX. Although the GR/PZ-AB/MT group exhibited a significantly higher bond strength than GR/UPA/RelyX, the GR/PZ-AB/SB group showed no significant difference relative to both the GR/PZ-AB/MT and GR/UPA/RelyX groups.

The normality ($P = 0.09$) and assumption of homoscedasticity ($P = 0.07$) corresponding to the results in Table 2 were substantiated. The results of two-way ANOVA indicated that bond strength was significantly influenced by the adhesive system type (No primer/SB, PZ-AB/SB, PZ-AB/MT, and UPA/RelyX; $P < 0.0001$) and the resin composite block material type (GR, SH, ES, and KZ; $P < 0.0001$), and that their interaction was significant ($P = 0.003$) (Table 3).

Failure mode

Most specimen failures fell into the Ad/Co category (Table 4). In addition, all specimens in the UPA/RelyX group exhibited the Ad failure mode, whereas three specimens in the GR/PZ-AB/MT group and one specimen each in the GR/PZ-AB/SB and KZ/PZ-AB/SB groups exhibited the Co failure mode. No specimen failed at the interface between the stainless-steel rod and the luting agent.

SEM observation

SEM images of the resin composite block materials are shown in Fig. 2. The GR contained irregularly shaped supra- and sub-micrometer particles surrounding the relatively large organic-inorganic composite filler. The SH exhibited spherical particles measuring $>1 \mu\text{m}$ in addition to indentations

where the particles had become detached from the matrix resin. The ES contained densely packed uniform spherical filler particles 100–200 nm in diameter with smooth surfaces. The KZ contained coarse filler particles measuring several hundred nanometers or less in diameter surrounded by a nano-porous structure.

Figure 3 shows the SH/PZ-AB/SB specimen, in which SH was bonded with the PZ-AB primer and the SB luting agent and showed Ad/Co mode failure. SEM observation of the debonded SH/PZ-AB/SB specimen showed that traces of fractured resin remained on the surface. Micrometer-sized concave surface features were observed, suggesting locations where filler particles had become detached from the surface.

Discussion

The tensile bond strength tests revealed that the bond strength was significantly influenced by the type of adhesive system and the resin composite block material (Table 3); therefore, the null hypothesis was rejected. The failure modes tended to shift from Ad to Co as the bond strength increased. The Co failure mode observed in the GR/PZ-AB/SB, KZ/PZ-AB/SB, and GR/PZ-AB/MT groups indicated that the adhesive force generated at the bonded interface was greater than the cohesive strength of the luting agents (Table 4).

The PZ-AB primer contained γ -MPTS with MDP, and all of the resin composite block materials used in this study contained a silica filler. Several studies have reported that MDP promotes bonding to alumina or zirconia [23–31]. Therefore, some chemical interactions were presumed to have occurred between MDP and the inorganic components of the substrate materials. In addition, γ -MPTS reacts with silica to form siloxane bonds and copolymerizes with methacrylates [32]. Acidic compounds activate the γ -MPTS, thereby accelerating the formation of siloxane bonds [33,34]. The effect of the PZ-AB primer could therefore be attributed to the cooperative effect of MDP and the γ -MPTS activated by the acidic MDP monomer.

Because the UPA/RelyX adhesive system also employed γ -MPTS, MDP, and MMA, this system was selected as a control. However, the KZ/UPA/RelyX, ES/UPA/RelyX, and SH/UPA/RelyX groups resulted in the lowest bond strengths. These findings suggest that no strong bonding is achieved without a suitable polymerization system.

Each of the resin composite block materials displayed different surface microstructures for their respective fillers, matrix resins, and interfaces (Fig. 2). The GR has an organic-inorganic composite filler; SH, a supra- and sub-micrometer spherical inorganic filler; ES, a uniform nano inorganic filler; and KZ, an aggregated nano inorganic filler, known as a

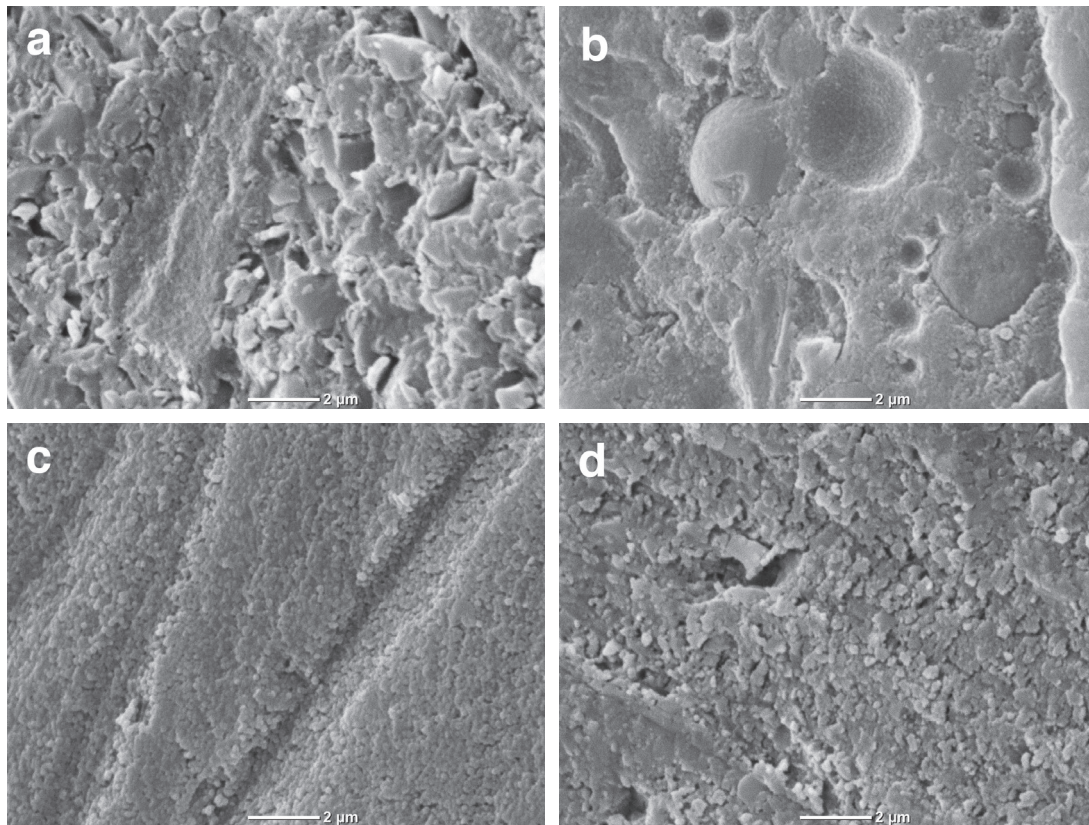


Fig. 2 Scanning electron microscopy images of resin composite block materials ground with a 600-grit silicon carbide abrasive paper: (a) Gradia Block (GR), (b) Shofu Block HC (SH), (c) Estelite Block (ES), (d) KZR-CAD HR2 (KZ) (original magnification $\times 8,000$)

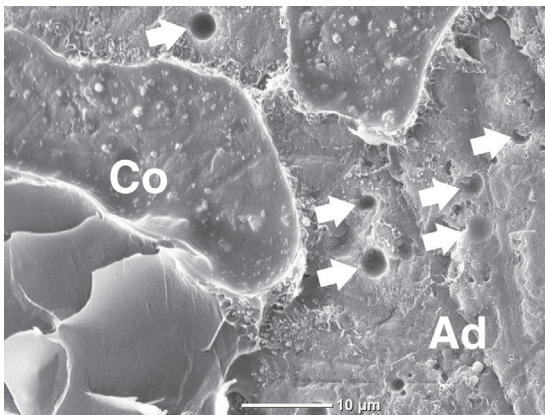


Fig. 3 The debonded surface of the SH specimen, which was bonded using the PZ-AB primer and the SB luting agent (original magnification $\times 2,000$). *Ad* and *Co* indicate adhesive failure at the luting agent-SH specimen interface and cohesive failure within the luting agent, respectively. Arrows indicate micro-convexities where filler particles had detached from the surface.

cluster filler. In contrast to GR, SH, and ES, the KZ exhibited relatively high bond strength when the SB resin was used without the primer. A characteristic feature of the cluster filler used in KZ is fusion of several sintered primary particles with diameters of 200-600 nm to form complex-shaped aggregates. As shown in Fig. 2, the ground surface of KZ is porous around the cluster filler, which is analogous to etched tooth enamel. As reported previously, the 4-META monomer of the SB resin promotes bonding by infiltrating the etched tooth surfaces [35]. Accordingly, it was speculated that the bonding mechanism involved MMA diffusion with 4-META into the narrow space that surrounds the cluster filler, followed by initiation of the polymerization process by TBB in the presence of water molecules [36] to generate micro-mechanical retention.

In the PZ-AB/SB group, GR, ES, and KZ exhibited significantly higher bond strengths than SH. However, in the UPA/RelyX group, GR resulted in superior bonding relative to SH, ES, and KZ. This may have resulted

from the presence of polymerizable C=C double bonds in GR [17], allowing chemical linkages to be formed with the other methacrylates through polymerization [37]. These findings suggest that, in order to maximize retention, clinicians should select an adhesive system suitable for the block material when fabricating resin composite restorations with CAD/CAM systems, particularly when limited mechanical retention exists on the abutment tooth.

Immediate bond strength development is essential to prevent restoration detachment caused by occlusal forces [19]. It is also useful for preclinical screening of different restorative materials and adhesive systems. Because the bond strength test performed in this study does not reflect all the factors present in the oral cavity, long-term clinical observations are required to validate the present findings. It has been reported that a luting agent producing a heavily crosslinked structure with a high elastic modulus is less durable against thermal stress than a linear polymer with a low elastic modulus, such as MT [38]. Therefore, the next research goal is to determine the relationship between bonding durability and the chemical components of various resin composite block materials using MMA-TBB-based resins.

In conclusion, it has been demonstrated that bond strength is significantly influenced by the type of adhesive system and the resin composite block material employed. MMA-TBB-based adhesive systems (PZ-AB/MT and PZ-AB/SB) exhibited higher bond strengths than the dual-curing composite-type adhesive system (UPA/RelyX) for each resin composite block material. For these adhesive systems, GR exhibited significantly higher bond strength than SH, whereas ES and KZ showed intermediate values.

Conflict of interest

The authors have no conflicts of interest to declare.

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