Journal of Materials Science and Engineering B 11 (1-3) (2021) 1-7
doi: 10.17265/2161-6221/2021.1-3.001

Effect of Universal Primers on the Tensile Bond Strength between Zirconia and Resin Composites

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Abstract: This study aimed to evaluate the effects of universal primers on the tensile bond strength between zirconia and resin composites. Zirconia specimens were divided into five groups based on the surface treatment with the following primers: MP (Monobond Plus), SU (ScotchBond Universal), AZ (AZ Primer), BM (Beauty bond Multi), and BL (Bondmer Lightless). After priming, stainless steel rods were bonded to the zirconia specimens with composite resin. The tensile bond strength test was performed: stored at room temperature for 1 day; stored in distilled water at 37 °C for 7 days; and underwent thermal cycling. The BL group demonstrated a significantly higher tensile bond strength than other groups when stored at room temperature for 1 day ($p < 0.05$). The primer that acted via chemical polymerization appeared to be most effective in improving the bond strength between the two materials in this study.

Key words: Universal primer, zirconia, composite resin, bond strength.

1. Introduction

The aesthetic demands of patients who receive dental treatment have demonstrated an increasing trend in recent years. The number of prosthetic treatments aimed at esthetic restoration is growing, and the widespread use of CAD/CAM (computer-aided design and computer-aided manufacturing) has made it possible to use various materials for esthetic treatment; for example, the use of ceramics, zirconia, and porcelain fused metal crowns is increasing [1]. It is necessary to use the appropriate primer as a pretreatment material for the adherend when bonding with a resin cement [2, 3]. However, when repairing a hard resin facing crown with a resin, it is necessary to specify the material and select the optimum primer. The selected primers have different processing times and drying methods; hence, they are associated with a risk of misoperation. It is important to control the expiration dates of different types of primers. Primers that are stored for long durations may present with a decrease in their adhesive strengths [4].

The restorative material is generally removed and a new restorative treatment is performed in cases where a fracture and secondary caries occur. This applies not only to prosthetic procedures but also to inlay and composite resin restorations. The adhesion of restorative materials to the tooth surface is becoming more reliable due to the advancements in dental adhesive technology. Restorative treatments using adhesives have gained popularity, in line with the concept of minimal intervention [5]. However, the problem arises when the different adherends are exposed.

Multi-purpose universal primers have been introduced to improve the speed, simplify the bonding process and reduce the chairside time in the clinical practice. These primers are capable of providing

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adhesive strengths to zirconia, ceramic, and precious metal adherends in a few steps. However, data on the strengths of these primes after long-term storage are currently unavailable, and the reliability of these materials, when used clinically, remains doubtful. In recent years, the price of precious metals has risen dramatically. There is a need for an alternative metal that can combine the workability of a precious metal and the strength of the bonding porcelain, and remain cost-effective. Currently, non-precious metal alloys using cobalt, chromium, and nickel are used; however, these materials might have a systemic effect, such as metal allergy [6]. Y-TZP (Yttria-stabilized tetragonal zirconia polycrystal) ceramic, a new type of ceramic has been attracting attention as an alternative material over the past few years [7]. Zirconia is a material with excellent biocompatibility [8] and aesthetics [9] and has sufficient mechanical strength. It is increasingly used as a long-term stable prosthetic restorative material. The purpose of the present study was to investigate the adhesive strengths of composite resins to zirconia using universal primers. The null hypothesis tested was that no significant differences would be found on bond strength produced by the different surface treatments tested among the materials stored at room temperature for 1 day or in water at 37 °C for 7 days or underwent thermal cycling .

2. Materials and Methods

2.1 Products Used

The universal primers used in this study were Beauty bond multi (BM; Shofu Inc., Kyoto, Japan; Lot. 021209), Monobond Plus (MP; Ivoclar Vivadent, Liechtenstein; Lot. S22510), Scotchbond Universal Adhesive (SU; 3M ESPE, Seefeld, Germany; Lot. 529681), and Bondmer Lightless (BL: Tokuyama, Dental Corporation, Tokyo, Japan; Lot. B170104). The AZ Primer (AZ; Shofu Inc., Kyoto, Japan; Lot. 101420) was used as a control for a total of five types of primer. The composition of the primer is described in Table 1.

2.2 Preparation of Test Specimens

A zirconia block (Y-TZP) was cut into pieces (20 × 20 × 3 mm) and polished with a water-resistant abrasive paper (grit, #400 and #600). The specimens underwent ultrasonic cleaning with acetone and distilled water and were dried at 50 °C for 2 h. After drying, various primers were applied to the zirconia surface using a microbrush, according to the manufacturers' instructions as follows: AZ was applied and left to dry naturally for 10 s; MP was applied and left to dry for 60 s followed by air-drying; BM was applied and left to dry for 10 s followed by air-drying for about 3 s (subsequently, the strength of the air was increased and light irradiation was performed for 10 s after the specimen had been sufficiently dried); SU was applied for 20 s, dried with weak air for 5 s, and then irradiated with light for 10 s; and in the case of BL, one drop each of solution A and solution B was sampled, mixed so that the solutions were uniform, applied thinly on the adherend, and dried sufficiently with air.

2.3 Evaluation of the Bond Strength

A mending tape (Scotch® Mending Tape 810, Sumitomo 3M, Tokyo, Japan) consisting of a hole (diameter, 3 mm) was attached to each surface-treated zirconia specimen to define the adhesion area. A chemical polymerization type composite resin (Clearfill FII, Clare Noritake Dental Co., Lot. 014012) was used for adhesion. About 0.2 g of the composite resin was placed on the adhesive surface of a mesh-type stainless steel rod 5 mm in diameter (Tomy International, Tokyo, Japan) and pressed under a load of 500 g for 1 min. After storage of the prepared specimens at room temperature for 24 h, they were divided into three groups as follows: one group was stored at room temperature for 1 day, one group was stored in distilled water at 37 °C for 7 days, and one group underwent thermalcycling (5,000×, 5 °C and 55 °C for 40 s). Each group was subjected to a tensile bond strength test using an autograph (EZ Test; Shimadzu,

Table 1 Used primer in this study.

Product	Manufacturer	Composition	Code
AZ Primer (Lot. 101420)	Shofu Inc., Kyoto, Japan	6-methacryloxyhexylphosphonoacetate, aceton, others	AZ
Beautibond multi (Lot. 021209)	Shofu Inc., Kyoto, Japan	Photo initiator, water, BiS-GMA (bisphenol A-glycidyl methacrylate), carboxylic acid monomer, TEGDMA (triethylene glycol dimethacrylate), phosphonic acid monomer, solvent	BM
Monobond Puls (Lot. S22510)	Ivoclar Vivadent Liechtenstein	MDP (10-methacryloyloxydecyldihydrogen phosphate), silane methacrylate, sufide methacrylate, ethanol	MP
Scotchbond Universal Adhesive (Lot. 529681)	3M Deutschaland GmbH Neuss, Germany	MDP, dimethacrylate resins, HEMA (2-hydroxyethyl methacrylate), vitrebond copoly, filer, ethanol, water, initiators, silane	SU
Bondmer lightless (Lot. B170104)	Tokuyama Dental Corp, Tokyou, Japan	Liquid A: phosphate monomer, BiS-GMA, TEGDMA, HEMA, MTU-6 (6-methacryloxyhexyl 2-thiouracil-5-carboxylate) Liquid B: acetone, isopropanol, water, acryl borate catalyst, γ-MPTS (methacryloxypropyl trimethoxysilane), peroxide	BL

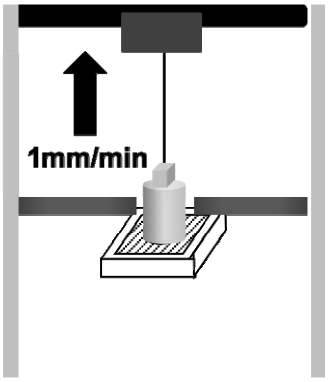


Fig. 1 Tensile bond test until rupture at a crosshead speed of 1 mm/min to determine the value at rupture.

Kyoto, Japan) at a crosshead speed of 1 mm/min (Fig. 1); five samples were used in each group. The fractured surfaces formed during the tensile bond strength test were observed using a stereomicroscope (GR1040-2S; Matsudensha, Tokyo, Japan). Three groups were formed according to the differences in the failure patterns: interfacial failure, due to the surface of zirconia; cohesive failure, due to the composite resin; and mixed failure, due to the failure at the interface between zirconia and the composite resin.

2.4 Contact Angle

The wettability of the zirconia specimens subjected to the various surface treatments was determined using an automatic contact angle meter (DCA-VZ, Kyowa Interface Science, Saitama, Japan). The measurement was performed using the drop method, wherein a 1.5 µL of resin monomer droplet was used (Bis-GMA:TEGDMA, 50:50). The contact angle value was calculated from the average of 10 measurement values.

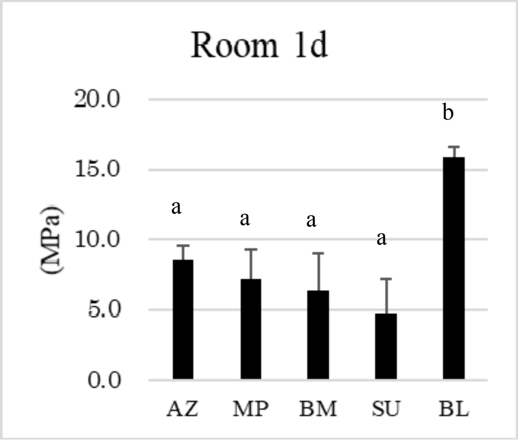
2.5 Statistical Analysis

The average values (± the standard deviations) were determined. Significant differences in mean values were assessed using one-way analysis of variance and Tukey's multiple comparison test. The significance level was set at $p < 0.05$. All statistical analyses were performed using the software SPSS (Statistical Product and Service Solutions), version 22.0 for Windows (SPSS Inc., Chicago, IL, USA).

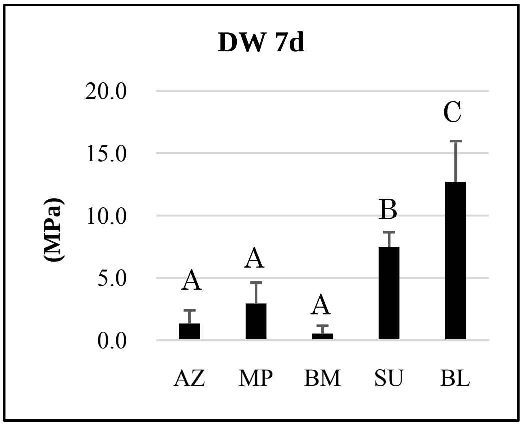
3. Results

3.1 Tensile Bond Strength

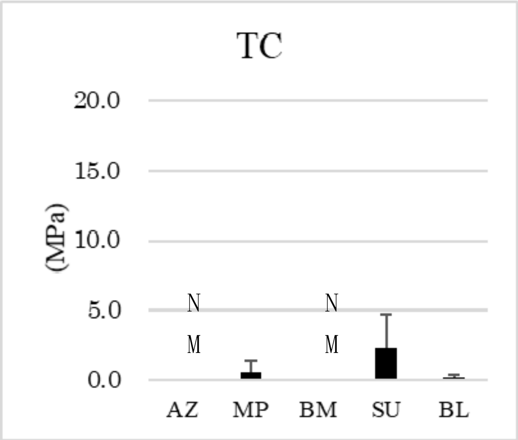
The results of the tensile bond strengths of the resin to the surfaces of the zirconia specimens are shown in Fig. 2. The tensile bond strength of BL was significantly higher initial bond strength than AZ, MP, BM and SU, when stored at room temperature for 1



Different superscript letters represent significant results ($p < 0.05$, $n = 5$, mean ± S.D.)



Different superscript letters represent significant results ($p < 0.05$, $n = 5$, mean ± S.D.)



NM = not measurable ($n = 5$, mean ± S.D.)

Fig. 2 Isolated comparison of mean bond strength (MPa) for each luting agent.

Room 1d: stored at room temperature for 1 day.

DW 7D: stored in distilled water at 37 °C for 7 days.

TC: underwent thermalcycling (5,000×, 5 °C and 55 °C for 40 s).

Different superscript letters represent significant results ($p < 0.05$).

day ($p < 0.05$). The tensile bond strength of SU was significantly higher bond strength than AZ, MP and BM when stored at room temperature for 7 days.

The tensile bond strength of BL was significantly higher bond strength than AZ, MP, BM and SU when stored at room temperature for 7 days. The samples of AZ and BM after thermocycles all de-bonded before testing. After thermocycles, all the primer surface treatments had no significant differences between in bond strength ($p < 0.05$).

3.2 Observation of Fractured Surface

The results related to the fractured surfaces after the tensile bond strength test are shown in Table 2. AZ demonstrated many interfacial failures after 1 day of storage at room temperature, and only interface failure was observed after 7 days of storage in water. MP demonstrated many cohesive failures after 1 day of storage at room temperature, and interface and mixed failure after 7 days of storage in water. BM demonstrated mixed and cohesive failures after 1 day of storage at room temperature, and only interface failure after 7 days of storage in water. SU demonstrated mixed and cohesive failures after 1 day of storage at room temperature, and many cohesive failures after 7 days of storage in water. Many cohesive failures were noted after storage for 1 day at room temperature in the case of BL, followed by mixed and cohesive failures after storage for 7 days in water. AZ, MP, BM and BL demonstrate only interface failure was observed after thermocycles.

3.3 Contact Angle (the Results of the Contact Angles of the Resin Monomers)

The results of the contact angles of the resin monomers following each surface treatment are shown in Table 3. The contact angles of AZ, MP, and BM were 19.2°, 18.7°, and 17.5° respectively; no significant differences were observed among the three materials. However, the contact angle of SU was significantly higher than those of AZ, BM, and MP at 24.0°($p < 0.05$).

Table 2 Failure modes according to tested groups.

	Room 1d			DW 7D			TC		
	IF	MF	CF	IF	MF	CF	IF	MF	CF
AZ	3	1	1	5	0	0	5	0	0
MP	1	1	3	2	3	0	5	0	0
BM	0	2	3	5	0	0	5	0	0
SU	0	2	3	0	1	4	2	2	1
BL	1	0	4	0	3	2	5	0	0

Room 1d: stored at room temperature for 1 day.

DW 7D: stored in distilled water at 37 °C for 7 days.

TC: underwent thermalcycling (5,000×, 5 °C and 55 °C for 40 s).

IF: interfacial failure; MF: mixed failure; CF: cohesive failure.

Table 3 The application effect of the primer of each adhesive system to the mirror-polished zirconia surface on the contact angle of a resin monomer drop.

	Contact angle of a resin monomer Mean ± S.D. (°)	
AZ	19.2 ± 1.6	a
MP	18.7 ± 0.6	a
BM	17.4 ± 0.8	a
SU	24.0 ± 3.2	b
BL	49.8 ± 2.2	c

There were 5 specimens in each experimental group.

Different superscript letters represent significant results ($p < 0.05$, $n = 5$).

BL presented with the highest contact angle 49.8°, which was significantly different from those of the other primers ($p < 0.05$; Table 3).

4. Discussion

The present study found that the surface treatment of the zirconia, the storage conditions affected the surface and the tensile bond strength between the zirconia and composite resin. Therefore, the null hypothesis was rejected.

Zirconia has been widely used to improve the aesthetics by the bonding of porcelain tozirconia [10]. However, occlusal forces such as bruxism may cause fracture at the interface between the two materials [11]. While repairing a fracture in a composite resin, it is possible to improve the adhesive strength by using a primer that is effective for each adherend. However, in cases where the facing crown is to be repaired, it is clinically difficult to identify the material used for the adherend and select the optimal primer. The universal primer can be used to overcome these issues.

On the other hand, sandblasting promotes the interlocking forces in adhesive materials and improves the bonding strength. The surface of the zirconia specimen is sandblasted to generate an activated fine surface, which increases the adhesive area, improves the wettability, and thereby, improves the adhesive strength [12, 13]. In the present study, we focused on the bond strength of the primer to zirconia and tested it without sandblasting.

The difference in the tensile bond strength test among the various primers was considered to be due to the differences in the compositions of the primers. It has been reported that MDP is effective as an adhesive monomer for zirconia [14-18]. The phosphate group in MDP can bond with the hydroxyl group in the oxide layer present in zirconia. The carboxylic acid group (methacrylate) has a carbon double bond, which can be polymerized and bonded to the composite resin. MP, SU, and BL contain phosphoric acid-based monomers; hence, they presented with less interfacial fractures when stored in water for 7 days. In particular, SU and BL showed significantly higher adhesion than AZ. BL is a two-liquid type monomer that contains Bis-GMA and TEGDMA, which are also found in composite resins, and the chemical polymerization is initiated by mixing the two liquids. It is thought that a strong resin layer is formed when this monomer is applied onto the surface of the zirconia specimen, which effectively acts on its adhesion to the composite resin. Furthermore, it is suggested that the borate-based catalyst promotes the polymerization of the composite resin [19]. In addition, it has been reported that the life of a single-bottle primer may be reduced due to the evaporation of its components thereby resulting in a reduction in the adhesive strength of the primer [20]. It has been suggested that a bottle of silane-coupling agent containing γ-MPTS and an acidic solution presented a reduction in the shelf-life of the solution due to the hydrolysis of γ-MPTS [21]. However, BL is a two-liquid type of monomer, wherein the γ-MPTS and MDP are stored in separate containers; the actions of the components are more effective because the reactions occur only after they are mixed [22].

In the thermal cycle durability test, a cycle of 5,000 times has been reported as the minimum number required for adhesion tests between metal and resin [23]. In the current study, all the primers presented low values after 5,000 thermal cycles indicating that chemical bonding with a primer alone was not effective for adhesion durability, as reported in a previous study [24]. Taken together, these findings and the results of the current study indicate that universal primers containing MDP are effective for the adhesion of a composite resin to zirconia. In particular, the two-liquid chemically-polymerized primer appeared to be most effective.

5. Conclusion

In this study, the adhesive strength between the zirconia surface and the composite resin was investigated using various multi-primers. The following conclusions were obtained:

(1) The adhesion durability was low and no significant difference in the surface modification of the zirconia specimen was observed among the various multi-primers.

(2) The tensile bond strength of BL was higher than those of the other multi-primers when the specimens were stored at room temperature for 1 day and in water for 7 days.

(3) The chemical polymerization reaction in two-bottle universal primers was found to be effective at the adhesive interface between the resin and the zirconia specimen.

Conflicts of Interest

There are no conflicts of interest to disclose in this study.

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