Original article

The effects of various surface treatments on the bond strength between lithium disilicate pressed-ceramics and luting agent

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Abstract

Purpose: This study aimed to evaluate the effects of various surface treatments, including resin coating, on the bond strength between lithium disilicate ceramics and luting agent.

Materials and Methods: All specimens were air-abraded with alumina particles and separated into the following four groups based on the surface treatment method used: silane coupling agent (PA); multipurpose ceramic/metal primer (CP); multipurpose ceramic/metal primer with resin coating (CM); and silica coating with multipurpose ceramic/metal primer followed by resin coating (RM). A luting agent was placed on the treated surface and photopolymerized. The shear bond strengths of the materials were measured after 10,000 thermal cycles.

Results: The bond strengths of the specimens in the PA group were significantly higher than those in the CP group. CM and RM groups presented with significantly higher bond strengths than CP.

Conclusion: The combination of multipurpose ceramic/metal primers and resin coating can significantly increase the bond strength between lithium disilicate ceramics and luting agent.

Key Words: adhesive bonding, lithium disilicate ceramics, resin coating, shear bond strength

Introduction

The demand for aesthetic prostheses is increasing, and lithium disilicate ceramics are widely used in clinical settings as a substitute for metal-ceramic restorations [1]. The prosthesis needs to firmly adhere to the abutment for the long-term stability of an all-ceramic restoration. The treatment of the inner surface of the prosthesis, which comes in contact with the abutment, considerably affects the long-term stability of the restoration. Treatment of the surfaces of silica-based ceramics using silane coupling agents has proven effective [2]. Multipurpose ceramic/metal primers, which are available as bottled mixtures of acidic functional monomers or water and a silane coupling agent, are widely used. However, the mixture is prone to dehydration synthesis and/or hydrolysis, which impairs the silanol activity because the monomers and agents can partially react within the bottle [3,4]. The resin-coating technique has been conventionally applied to abutments. Previous studies have shown that the resin-coating technique can be effectively used for bonding in indirect restorations [5,6]. Therefore, the aim of this study was to investigate the use of a silane coupling agent, activated immediately before application, along with a widely used multipurpose ceramic/metal primer, with and without the application of a resin coating on the inner surface of a prosthesis. The effects of the surface treatments on the bond durability between the lithium disilicate ceramics and the luting agent were evaluated.

Materials and Methods

Bond strength test

Preparation of specimen

The materials used in this study are shown in Table 1. The experimental design used in this study is outlined in Fig. 1. A low-speed precision cutting machine (Isomet, Buehler, Chicago, IL, USA) was used to excise 36 specimens (diameter, 13 mm; thickness, 5 mm) from a lithium disilicate ceramic block. The specimens were polished with a waterproof abrasive paper (grit 600; Sankyo Rikagaku, Okegawa, Japan) and embedded in autopolymerizing resin (Ostron II, GC, Tokyo, Japan; Fig. 1a, b). The adherend surfaces were air-abraded with alumina particles (50 μm) at 0.3 MPa pressure for 15 s (working distance, 10 mm) followed by rinsing in distilled water and drying by air blowing. The adherend surface areas on the lithium disilicate ceramics were defined using masking tape. The 36 specimens were divided into four groups (n = 9, each) based on the surface treatment provided as follows (Fig. 1c):

1. Silane coupling agent group (PA), where a silane coupling agent (Clearfil Porcelain Bond Activator, Kuraray Noritake Dental, Tokyo, Japan) was mixed with a self-etching primer (Clearfil Megabond 2 Primer, Kuraray Noritake) and applied to the surface.
2. Multipurpose ceramic/metal primer group (CP), where a multipurpose ceramic/metal primer (Ceramic Primer Plus, Kuraray Noritake Dental) was applied onto the surface.
3. Multipurpose ceramic/metal primer with resin coating group (CM), where a resin coating was applied to the surface after applying the multipurpose ceramic/metal primer (Ceramic Primer Plus). The light-cure bonding agent (Clearfil
Megabond 2 Bond, Kuraray Noritake Dental) was applied to the surface, according to the manufacturer’s instructions. After air blowing, the surface was photopolymerized using an LED curing light unit (Elitedent Q-4, Rolence, Chungli, Taoyuan, Taiwan, ROC) for 30 s at an irradiance of 1,400 mW/cm².

(4) Silica coating with multipurpose ceramic/metal primer followed by resin coating group (RM), wherein the surface was coated with silica-coated alumina particles (30 μm; Rocatec Soft, 3M ESPE, Seefeld, Germany) and a resin coating was applied after the application of the multipurpose ceramic/metal primer.

After completing the surface treatments, the amount of cement was standardized by attaching an aluminum pipe (internal diameter, 5.0 mm; height, 2.0 mm) to the adherend using sticky wax. The luting agent (Panavia V5, Kuraray Noritake Dental) was incrementally placed into the aluminum pipe and photopolymerized for 20 s using the LED light-curing unit (Fig. 1d).

### Table 1 Materials used in this study

| Brand name                  | Composition                                                                 | Batch no. | Manufacturer                  |
|-----------------------------|-----------------------------------------------------------------------------|-----------|------------------------------|
| IPS e.max Press             | SiO₂, Li₂O, K₂O, P₂O₅, ZrO₂, ZnO, other oxides, ceramic pigments            | X27174    | Ivoclar-Vivadent, Zurich, Switzerland |
| Panavia V5 Paste            | Bis-GMA, TEGDMA, hydrophobic aromatic dimethacrylate, hydrophilic aliphatic dimethacrylate, initiators, accelerators, silanated barium glass filler, fluoroaluminosilicate glass filler, colloidal silica, silanated aluminum oxide filler, dl-camphorquinone, pigments | 7V0037    | Kuraray Noritake Dental, Tokyo, Japan |
| Clearfil Porcelain Bond Activator | TMSPMA, hydrophobic aromatic dimethacrylate                           | 8A0032    | Kuraray Noritake Dental |
| Clearfil Megabond 2 Primer  | MDP, HEMA, hydrophilic aliphatic dimethacrylate, dl-camphorquinone, water  | 5R0044    | Kuraray Noritake Dental |
| Ceramic Primer Plus         | TMSPMA MDP, ethanol                                                        | 7R0018    | Kuraray Noritake Dental |
| Rocatec Soft                | 30 μm silica-coated alumina particles                                      | 016       | 3M ESPE, Seefeld, Germany     |
| Clearfil Megabond 2 Bond    | MDP, Bis-GMA, HEMA, hydrophobic aliphatic dimethacrylate, dl-camphorquinone, initiators, accelerators, silanated colloidal silica | 5T0062    | Kuraray Noritake Dental |

Bis-GMA, bis-phenol-A-diglycidylmethacrylate; TEGDMA, triethylene glycol dimethacrylate; TMSPMA, 3-trimethoxysilylpropyl methacrylate; MDP, 10-methacryloyloxydecyl dihydrogen phosphate; HEMA, 2-hydroxyethyl methacrylate

### Specimen storage

The nine specimens from each surface treatment group were stored at room temperature for 24 h. Subsequently, they were subjected to a thermal cycle aging test (10,000 cycles; 5 to 55°C; dwell time, 40 s) to assess the bonding durability (Fig. 1e).

### Shear bond strength measurement

After bonding and storing the specimens, the shear bond strength was tested using a universal testing machine (EZ Test, Shimadzu, Kyoto, Japan) at a crosshead speed of 0.5 mm/min (Fig. 1f).

### Observation of the treated surfaces

The specimens were prepared by cutting a lithium disilicate ceramic block with a low-speed precision cutting machine, polishing with waterproof abrasive paper (grit #600), and subjecting them to one of the following three surface treatments: polishing the surface with waterproof abrasive paper (#600); alumina air-abrasion of the surface, based on the fabrication of the specimen for the shear bond strength test; and silica coating of the surface after alumina air-abrasion, according to the fabrication of the specimen in Group RM.

The surfaces of the lithium disilicate ceramic specimens were observed under an electron microscope (JCM-6000Plus, JEOL, Akishima, Japan) at an accelerating voltage of 10 kV and magnifications of ×800 and ×3,000.

### Statistical analysis

Kolmogorov-Smirnov test (p = 0.200) for each measurement showed normal distribution in all experimental groups. Levene’s test of equality of variance was performed to assess the equality of variances in the shear bond strength of all experimental groups. The variance through the groups was found to be unequal (p = 0.001); therefore, non-
parametric tests were used to analyze the data in this study. The shear bond strength data were subjected to the Kruskal-Wallis test followed by the Dunn-Bonferroni method (significance level, $\alpha = 0.05$). SPSS (Ver. 25 for Windows, SPSS Inc., Chicago, IL, USA) was used for the statistical analysis.

(a) Lithium disilicate ceramics  
(b) Lithium disilicate ceramic discs were embedded in autopolymerizing resin  
(c) Surface treatment  
(d) Luting agent was injected into the mold + Light curing  
(e) 24-h storage at room temperature + Thermocycling test  
(f) Shear bond strength test

Fig. 1 Experimental design

Results

The results of the shear bond strength tests are summarized in Table 2 and Fig. 2. After the thermal cycles, the bond strengths of the specimens in the group treated with multipurpose ceramic/metal primer (CP; 2.5 ± 0.9 MPa) were significantly lower ($p < 0.05$) than those in the silane coupling agent group (PA; 5.0 ± 1.2 MPa).

The shear bond strengths of the two resin-coated groups (CM and RM; 5.4 ± 2.6 MPa and 6.6 ± 1.3 MPa, respectively) were significantly higher than that of the CP group (2.5 ± 0.9 MPa). On the other hand, no significant differences in shear bond strength were observed between the PA group and the CM and RM groups. Furthermore, no significant differences in bond strength were observed between the CM and RM groups.

Interfacial failure was the failure mode observed between the luting agent and lithium disilicate ceramics in all four groups. In the surface observations, the polished surface appeared smooth with little unevenness (Fig. 3a). The surface that was air-abraded with alumina particles appeared rough and irregular (Fig. 3b), whereas the silica-coated surface was irregular and uneven with consisted of fine particles attached to it (Fig. 3c).

Table 2 Shear bond strengths of the luting agent to lithium disilicate pressed-ceramics

| Surface treatment | PA      | CP      | CM      | RM      |
|-------------------|---------|---------|---------|---------|
| Shear bond strength (MPa) | 5.0 (1.2)$^A$ | 2.5 (0.9)$^B$ | 5.4 (2.6)$^A$ | 6.6 (1.3)$^A$ |

Bond strengths mean and standard deviation in parentheses. Significant differences are visualized with different letters.
Discussion

Multipurpose ceramic/metal primers are widely used to treat the surfaces of all kinds of materials, including ceramics and metals. However, they are prone to dehydration condensation and/or deterioration of the silane coupling agent component (3-trimethoxysilylpropyl methacrylate, TMSPMA) because the acidic functional monomers and silane coupling agent exist within the same solution [3]. Yoshikawa et al. reported that the use of a TMSPMA silane coupling agent immediately after mixing with multipurpose a ceramic/metal primer resulted in a significant increase in the bond strength when compared to the use of a multipurpose ceramic/metal primer alone. On the other hand, a decrease in bond strength was observed when a mixture of a multipurpose ceramic/metal primer and a TMSPMA silane coupling agent was kept for one day or more before use; it was suspected that the silane coupling agent in the multipurpose ceramic/metal primer had partially lost its activity before use [4]. These findings were corroborated by the results of the shear bond strength measurements in the current study. The silane coupling agent that was activated immediately before use maintained long-term stability, whereas the commonly used multipurpose ceramic/metal primer showed lowered activity before use, indicating a decrease in the long-term durability. The bond strengths of the two resin-coated groups were significantly higher than that of the multipurpose ceramic/metal primer group, indicating that the combination of a resin coating with a multipurpose ceramic/metal primer can increase the long-term durability of the material.

Surfaces that are silica-coated using Rocatec manifest a unique foamy structure that is different from the surfaces that are air-abraded with alumina particles. The foamy structure is believed to form a fractal structure and increase the wettability of the surface [7]. In the scanning electron microscopic (SEM) observations in the current study, fine particles related to this foamy structure were observed on the silica-coated surfaces. However, the bond strength of the RM group, which was treated with a silica coating and a multipurpose ceramic/metal primer followed by a resin coating, was higher than that of the CM group, which was treated with a multipurpose ceramic/metal primer and a resin coating, statistical significance notwithstanding.

Tribochemical silica coating has been reported to be effective in improving the bond strength between non-silicate ceramics, such as aluminum-oxide ceramics and zirconia-dioxide ceramics, and resin cements [8]. Lithium disilicate ceramics has a high flexural strength, which is achieved by embedding lithium disilicate crystals in a glass matrix [9,10], and contains silicon dioxide. The results of the present study suggest that the effect of silica coating is possibly low in lithium disilicate ceramics compared to that in non-silicate ceramics because the former contains silicon dioxide on the adherend surface.

Erdemir et al. reported that tribochemical silica coating was effective in repairing lithium disilicate ceramics using resin [11]. They used a silane coupling agent that did not contain acidic functional monomers. Alternatively, the multipurpose ceramic/metal primer used in the current study contained acidic functional monomers, which might have lowered the activity of the TMSPMA silane coupling agent and resulted in the ineffectiveness of the tribochemical silica coating.

Etching with hydrofluoric acid is believed to improve the wettability of the silicate ceramic surface [12] and strengthen the bond to the resin cement [13,14,15,16]. Similarly, the bond strength between lithium disilicate ceramics and resin cement has been reported to increase by treating the surface with hydrofluoric acid and a silane coupling agent [17,18,19,20]. However, in Japan, hydrofluoric acid is not commonly used as surface treatment, due to the fatal accident caused by misuse of hydrofluoric acid in dental clinic. Therefore, in this study, the adherend surfaces were air-abraded with alumina particles, instead of etching with hydrofluoric acid.

Hydrofluoric acid produces toxic gas; hence, special precautions are necessary when using the chemical. A self-etching glass-ceramic primer was developed by removing the toxic components from hydrofluoric acid. It can perform both etching and silanization at the same time and is reported to achieve satisfactory bond strength [21]. However, there are
reports on the poor bond strength of the self-etching glass-ceramic primer when compared to that achieved with hydrofluoric acid and a silane coupling agent [18,19], which might be attributed to the deterioration in the activity of the silane coupling agent because it was contained in the same bottle with hydrofluoric acid, as in the case of the multipurpose ceramic/metal primers (acidic functional monomers and a silane agent are contained in the same bottle).

In this study, the resin-coating technique [5,6] was applied to the inner side of the prosthesis. Lithium disilicate ceramic specimens were treated with a multipurpose ceramic/metal primer, on which the bonding agent was applied, and the surface was photopolymerized. This procedure is associated with a risk of the prosthesis floating from the abutment, which can be prevented by adding the appropriate cement space. A cement space of 120 μm or less is considered to be clinically acceptable [22]. Furthermore, according to Diaz-Arnold et al., the highest bond strength of a resin cement was obtained with a cement space of 80 μm [23]. In a recent study by Iwashita et al, the thickness of the coat formed by resin coating was 12.66 μm [24]. Therefore, the thickness of the resin coat is less likely to exceed the appropriate cement space or affect the fitting precision. Moreover, recent advancements in digital technology have led to the use of a prosthesis that is manufactured via computer-aided design and manufacturing (CAD/CAM) [1], which can add arbitrary cement space; thus, the problem of floating can be prevented by designing the prosthesis based on the thickness of the resin coat. These findings indicate that the application of a resin coating on lithium disilicate ceramics is a simple, safe, and effective method to increase its bond strength to a luting agent.

In this study, the effects of various surface treatments, including resin coating, were evaluated on the bond strength between lithium disilicate ceramics and luting agent. A multipurpose ceramic/metal primer can undergo deterioration, such as hydrolysis, inside the bottle. Hence, it is desirable to use a silane agent that is activated immediately before use. The combination of a multipurpose ceramic/metal primer and resin coating can significantly increase the bond strength between lithium disilicate ceramics and luting agent.

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Conflicts of Interest
None

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