Combination of a silane coupling agent and resin primer reinforces bonding effectiveness to a CAD/CAM indirect resin composite block

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The effects of silanization and resin primer application on CAD/CAM indirect resin composite block bonding were investigated. KATANA AVENCIA P blocks (Kuraray Noritake Dental) were treated with a silane coupling agent and/or a resin primer. The contact angles (CAs) of resin primer were observed before and after silanization. Panavia V5 (Kuraray Noritake Dental) was built after each treatment. Bond strengths were measured, and the interface was analyzed by energy dispersive X-ray spectroscopy (EDS). The CA showed that silane treatment improved the wettability of the resin primer to the resin block. The combination treatment of the silane and resin primer showed significantly higher bond strength than no treatment, only in the silanization or resin primer group (p<0.001 each). EDS analysis showed that the resin primer penetrated both cement and block sides. The combination of the silane and resin primer improved bonding effectiveness between the resin block and resin cement.

Keywords: Adhesive dentistry, Dental bonding, Longevity, Micro-tensile bond strength, Spectrometry/X-ray emission

INTRODUCTION

Computer-aided design/computer-aided manufacturing (CAD/CAM) systems have become widely adopted in dental treatment, and they allow the use of several metal-free materials, such as zirconia, and composite resin1-2. Composite resin blocks that consist of a glass phase and a matrix resin phase provide an esthetic material whose properties are achieved by control of temperature and pressure, increasing the compounding amount of filler, and high levels of polymerization; consequently, these blocks are considered useful for crowns and inlays6. It has been previously reported that the mechanical properties and bond strength of CAD/CAM resin blocks are higher than those of glass ceramics3-5. In Japan, the Japanese social insurance system approved indirect composite premolar crowns prepared by CAD/CAM technology for restoration in 20146. Though there is a rapidly increasing demand for these blocks, investigation of suitable pre-surface treatments is ongoing. Several studies have examined surface treatments, such as airborne-particle abrasion and chemical treatment, to determine which is appropriate7-9. Currently, silane treatment after blasting is recommended as a pretreatment for bonding CAD/CAM resin blocks7,9-11. The bonding effectiveness is improved by blasting with alumina particles to strengthen the mechanical connection10,11 and by increasing the chemical interaction with the inorganic filler by silane treatment12-15. γ-methacryloxypropyltrimethoxysilane (γ-MPTS) is widely used as a silane coupling agent16,17, and it is activated in an acidic environment to form silanol groups and then binding to inorganic substrates such as fillers16,18. However, the activated silane coupling agents are chemically unstable substances, and it has been reported that their bonding ability decreases within a short time19,20. In addition, it is possible that the activity of the silane treatment has been lost by the time the bonding treatment is performed due to the molecular structure of the silane coupling agent, the temperature and humidity during primer storage, and the effect of the solvent15,20,21. The effect of silanization depends on: (1) silane concentration; (2) silane functional groups (matching with the resin monomer functional groups); (3) blending with cross-linking silanes; (4) pH; (5) hydrolysis time; (6) temperature; and (7) nature of the solvent mixture18. It has also been reported that even after the bonding treatment, the siloxane bond is hydrolyzed by the water present at the interface, reducing the bond strength22,23. In addition, bond degradation over time in the oral cavity is still the major concern18. These problems of these silane treatments need to be resolved in order to improve the clinical results of CAD/CAM resin crowns.

Since the CAD/CAM resin block is a composite of filler and matrix resin, not only filler, the matrix resin is also exposed on the surface. Reymus et al. reported that a resin primer treatment to the matrix resin showed...
higher bond strength than a silane treatment for CAD/CAM resin block bonding\textsuperscript{24}. Fujii \textit{et al.} also reported the bonding performance of resin primer containing methyl methacrylate (MMA) in past studies\textsuperscript{25}. In our previous study\textsuperscript{26}, the MMA containing resin primer treatment showed higher long-term bond strength than the silane treatment. Since the resin primer treatment targets the matrix resin, it is possible that even higher bond strength can be achieved by using the resin primer treatment in combination with the silane treatment that targets the filler as the bonding target. However, no report has examined the effect of both silane treatment and MMA-containing primer treatment on the CAD/CAM resin block in detail. Therefore, the aim of this study was to clarify the effect of both MMA-containing primer and silane treatments on long-term adhesion to the CAD/CAM resin block. The null hypothesis was that MMA-containing primer treatment after silane treatment does not affect the bond strength between the CAD/CAM resin block and the adhesive resin cement.

**MATERIALS AND METHODS**

**Specimen preparation**

Three CAD/CAM resin blocks (KATANA AVENCIA P block, A3-LT, Kuraray Noritake Dental, Tokyo, Japan) were cut to the size of 7×7×4 mm using a diamond cutter (MC201-N, Maruto, Tokyo, Japan) (Table 1). Silicon carbide paper (PRO-ACT, 400 grit, KOHNAN, Osaka, Japan) was used to wet polish the surface (Fig. 1a). The block surfaces were then blasted with alumina (particle size was 50 µm) using a dental abrasion device (Adabrader, Morita, Tokyo, Japan) for 15 s, working at 10 mm from the surface of the block with a pressure of 0.2 MPa. A syringe was then used to dry the block surfaces by air flow for 10 s (Fig. 1b). These pretreated specimens were then used in four different trials (n=3 blocks per trial). Combinations of primer containing MMA (HC primer, Shofu, Kyoto, Japan) and a silane coupling agent (Clearfil Ceramic Primer Plus, Kuraray Noritake Dental) were then tested in four different adhesive protocols: (1) no adhesive, representing the Control group; (2) the HC primer was applied to the adhesive surface over a time span of 10 s and dried for 10 s, representing the Primer group; (3) the Clearfil Ceramic Primer Plus was applied to the adhesive surface over a time span of 10 s and dried for 10 s, representing the Silane group; and (4) the Clearfil Ceramic Primer Plus was applied to the adhesive surface over a time span of 10 s and dried for 10 s. Then, the HC primer was applied to the adhesive surface over a time span of 10 s and dried for 10 s, representing the Silane+Primer group (Fig. 1c).

**Micro-tensile bond strength (µTBS) tests**

The composite resin cement (Panavia V5, Kuraray Noritake Dental) was filled in 2-mm layers to a height of 4 mm on the surface after each treatment (Fig. 1d). The cement was light-cured using a light emitting diode (SATELEC Mini LED III, Acteon, Merignac, France) with a peak power output of 2,200 mW/cm² for 20 s (Fig. 1e). The specimens were then stored for 24 h in water at 37°C (Fig. 1f), after which a slow-speed, water-cooled, diamond saw (MC-201N, Maruto) was used to cut each specimen into beams measuring 0.7×0.7 mm (24 beams per block, total 72 beams) (Fig. 1g). Then, for the initial µTBS testing after 24 h (0M), 24 beams were selected at random (Fig. 1h). Using cyanoacrylate glue (Model Repair II Blue, Dentsply Sirona, Tokyo, Japan),

**Table 1 Materials**

| Material                      | Product (manufacturer), [lot number] | Composition                                                                 |
|-------------------------------|-------------------------------------|----------------------------------------------------------------------------|
| CAD/CAM resin block           | KATANA AVENCIA P block (Kuraray Noritake Dental), [171121] | UDMA, TEGDMA, silica powder, fine particulate silica, zirconium silicate, colorant |
| Surface treatment agent       | Clearfil Ceramic Primer Plus (Kuraray Noritake Dental), [A90009] | Ethanol, 3-trimethoxysilylpropyl methacrylate, 10-Methacryloyloxydecyl dihydrogen phosphate |
|                               | HC primer (Shofu), [031601]         | UDMA, MMA, polymerization initiator, acetone                               |
| Composite resin cement        | PANAVIA V5 (Kuraray Noritake Dental), [A60064] | Paste A: Bis-GMA, TEGDMA, hydrophobic aromatic dimethacrylate, hydrophilic aliphatic dimethacrylate, initiators, accelerators, silanated barium glass filler, silanated fluoroaluminosilicate glass filler, colloidal silica Paste B: Bis-GMA, hydrophobic aromatic dimethacrylate, hydrophilic aliphatic dimethacrylate, silanated barium glass filler, silanated aluminum oxide filler, accelerators, di-Camphorquinone, pigments |

UDMA: urethane dimethacrylate, TEGDMA: triethyleneglycol dimethacrylate, MMA: methyl methacrylate, Bis-GMA: bisphenol A-glycidyl methacrylate
each beam was fixed to a jig to subject it to a tensile force at a crosshead speed of 1.0 mm/min in a universal testing machine (EZtest, Shimadzu, Kyoto, Japan) until fracture. Mean and standard deviation values were then calculated (as MPa). As for the remaining beams, they were randomly divided into two groups to measure µTBS after 1 month (1M) or after 6 months (6M). For the 6-month group, the specimens were immersed and kept in the same water at 37°C in containers that were completely sealed.

Fracture mode distributions
The fracture surfaces after the µTBS measurements were analyzed using optical microscopy (30× magnification). Failure modes were classified into three modes: mixed failure (including adhesive failure) and cohesive failure within the CAD/CAM resin block or the composite resin cement.

Contact angle (CA) measurement
To assess the wettability of the MMA-containing primer on the surface of the block with different surface treatments, CA measurements were performed. CAD/CAM resin blocks (KATANA AVENCIA P block, A3-LT) were cut to the size of 7×7×4 mm using a diamond cutter (MC201N, Maruto). The Control group and the Silane group were prepared in the same manner as for the µTBS tests. A fully automatic small CA meter (P200A, Meiwaforsys, Tokyo, Japan) was used to measure CAs in a temperature-controlled room at 23±1°C with relative humidity of 50±5%. Using the dynamic sessile drop method, 9 drops of HC primer with a volume of 1 µL each were evaluated immediately after dropping, and the results were averaged for the analysis (Fig. 1i).

Element analysis
Testing specimens were prepared in the same manner as for the µTBS tests, and they were cut at 2-mm thickness to cross the interface with a diamond cutter (MC201-N, Maruto) (Fig. 1j). Each specimen was osmium-deposited by an OSMIUM PLASMA COATER (OPC60A, Filgen, Nagoya, Japan). Then, these were examined using FE-SEM (JSM-6335F, JEOL, Tokyo, Japan) equipped with energy dispersive X-ray spectroscopy (EDS; JED-2300, JEOL) (Fig. 1k). X-ray analysis was performed at an accelerating voltage of 20.0 kV (n=1 in each group).

Statistical analyses
All data were checked for normality and homoscedasticity by the Kolmogorov-Smirnov test and Bartlett’s test. The Kruskal-Wallis test and Mann-Whitney U test with Bonferroni correction were used to analyze the µTBS data, and Student’s t-test was used to analyze the CA data. The level of statistical significance was set at 5%. All statistical analyses were carried out with EZR software (www.jichi.ac.jp/saitama-sct/SaitamaHP.files/statmed.html).

RESULTS
µTBS tests
The Kruskal-Wallis test results showed that ‘surface treatment’ (p<0.001) and ‘water aging’ (p<0.001) had significant effects on the µTBS values (Table 2, Fig. 2). On multiple comparison testing, the Silane+Primer group showed significantly higher bond strengths than the Control group, the Primer group, and the Silane group (vs. Control: p<0.001, vs. Primer: p<0.001, vs.
Table 2  µTBS values (MPa) and failure modes

|       | Cont            | Primer          | Silane          | Silane + Primer |
|-------|-----------------|-----------------|-----------------|-----------------|
| 0M    | 71.59 (7.41) [22/2/0] | 73.33 (6.64) [20/4/0] | 84.17 (4.50) [23/1/0] | 100.88 (7.49) [7/17/0] |
| 1M    | 57.02 (6.73) [24/0/0] | 55.12 (6.27) [22/2/0] | 58.07 (5.74) [23/1/0] | 73.48 (10.89) [3/21/0] |
| 6M    | 54.52 (7.91) [21/3/0] | 55.38 (6.40) [23/1/0] | 52.94 (5.59) [24/0/0] | 73.57 (13.51) [9/15/0] |

Data are shown as mean (SD), n=24/group. Numbers in square brackets are the number of specimens classified into three modes [A/B/C]: [A] adhesive and mixed failure along the resin block-composite resin cement interface; [B] cohesive failure within the CAD/CAM resin block; [C] cohesive failure within the composite resin cement. 0M: 24 h of water storage, 1M: 1 month of water storage, 6M: 6 months of water storage.

Silane: *p<0.001). On the other hand, no significant difference was observed in the bond strengths among the Control group, the Primer group, and the Silane group (Control vs. Primer: p=0.99, Control vs. Silane group: p=0.87, Primer vs. Silane: p=0.99). In addition, the bond strengths decreased significantly in the first month, and there was no significant difference after 1 month (0M vs. 1M: p<0.001, 0M vs. 6M: p<0.001, 1M vs. 6M: p=0.43).

Failure mode analysis
In the Control group, the Primer group, and the Silane group, mixed failures were observed (Fig. 3). On the other hand, cohesive failures in the resin cement were observed more frequently in the Silane+Primer group. In all groups, no effect of storage time in water on fracture morphology was observed.

CA measurement
Student’s t-test showed that the Silane group (20.1±1.45°) had a significantly smaller CA than the Control group (23.9±5.08°; p=0.049, Fig. 4).

EDS analysis
On EDS analysis, filler components such as silicon (Si), aluminum (Al), barium (Ba), and oxygen (O) were found in the CAD/CAM resin block and resin cement (Fig. 5). In contrast, a large amount of carbon (C), which was abundant in matrix resins and MMA-containing primers, was detected. Elemental mapping analysis at the adhesive interface showed dense carbon accumulation along the interface (Fig. 5, white arrow). In addition, in the image in which the carbon mapping image was superimposed on the SEM observation image, an integrated image of carbon was detected near the bonding surface of the CAD/CAM resin block of the Primer and the Silane+Primer groups (Fig. 6). In contrast, no carbon-dense layer was observed in the Control group and the Silane group, and a clear interface was observed along the bonding surface.

In the carbon mapping image after each treatment, in both the Primer group and the Silane+Primer group coated with the MMA-containing primer, the carbon-dense layer of about 5 µm containing a filler was also found near the interface (Fig. 7). In addition, on
Fig. 5  SEM/EDS mapping images of adhesive surfaces of the Silane+Primer group.
(a) SEM: A higher density image of filler is observed on the block than on the resin cement. A tight interaction of block and cement is observed. C: carbon mapping, where a higher carbon density image is observed on the cement than on the block. Elemental mapping analysis shows a carbon-dense layer at the adhesive interface (white arrow). Si: silicon mapping, where a higher silicon density image is observed on the block than on the cement. O: oxygen mapping. Al: aluminum mapping. Ba: barium mapping. Bl: CAD/CAM resin block. Ce: Composite resin cement. (b) SEM: A rough block surface by alumina blasting is observed (black arrow head). C: carbon mapping, where a carbon-dense layer of 5 µm is observed (white arrow).

Fig. 6  SEM images and EDS carbon mapping of adhesive surfaces.
In the Primer group and the Silane+Primer group, the carbon-dense layer is observed in the carbon mapping image (white arrows). Bl: CAD/CAM resin block. Ce: Composite resin cement.

Fig. 7  Carbon and Silicon EDS mapping images of adhesive surfaces and elemental liner analysis in the Primer group and the Silane+Primer group. The peak of carbon decreases transitionally from the bonding interface to the resin cement. The carbon-dense layer of the Silane+Primer group is thinner than that of the Primer group. In the case of silicon, in contrast to carbon, the distribution ratio of silicon increases from the bonding interface toward the resin cement.
elemental linear analysis of carbon and silicon, the peak of carbon decreased transitonally from the bonding interface to the resin cement, showing that the carbon-dense layer was thin. On the other hand, in the case of silicon, in contrast to carbon, the distribution ratio of silicon increased from the bonding interface toward the resin cement.

**DISCUSSION**

The resin primer is defined as the primer for which the silane coupling agent is not included, and the matrix resin is the target for bonding. The silane treatment and the resin primer treatment have different adhesion targets on the block adhesion surface (i.e., filler or matrix resin), so it is possible that higher adhesion can be obtained by performing both the silane treatment and the resin primer treatment. In this study, the only commercially available resin primer for CAD/CAM resin crown bonding (i.e., MMA-containing primer) was used. On the μTBS test, the Silane+Primer group showed significantly higher bond strength than the Control group, the Silane group, and the Primer group. Therefore, the null hypothesis, MMA-containing primer treatment after silane treatment does not affect the bond strength between the CAD/CAM resin block and the adhesive resin cement, was rejected. The Silane+Primer group showed higher bond strength than the other groups not only after 24 h, but also after 6 months of water storage, and cohesive failure within the resin cement was observed. It was found that the degradation of the interface was less likely to occur than in the other groups. Fujii et al. also reported that higher bond strength was obtained when MMA-containing primer treatment and silane treatment were used in combination compared with silane treatment alone, but the results were only for initial bond strength (i.e., after 24-h water storage). The present study also showed the long-term durability of the silane and MMA-containing primer treatment. Moreover, the synergistic effect of silane treatment and primer treatment was clarified for the first time based on wettability and elemental analysis of the adhesive interface using blocks with a high filler content.

MMA-containing treatment alone did not affect the bonding effectiveness in the present study, even though the carbon-dense layer was observed by EDS (Figs. 6, 7). In our previous study, the MMA-containing primers improved the initial bond strength to the resin block. The previous study examined the MMA-containing primer (HC primer, i.e., same as the present study), but the resin composite block (SHOFU BLOCK HC, Shofu) that was used was different. In the Japanese social insurance system, SHOFU BLOCK HC is used for premolars, and KATANA AVENCIA P block containing higher amount of filler is used for molars. Theoretically, if the amount of filler in the resin composite increases, the matrix ratio will decrease, possibly decreasing the effect of the resin primer. This can be the reason for the discrepancy between the previous study’s results and those of the present study.

Silane treatment alone did not affect the μTBS, and the interface between resin block and cement was not different from that seen in the Control group under SEM/EDS observation. Resin composites consist of a matrix resin and various fillers, including glass ceramics or dental glasses, and silane coupling agents can provide a suitably strong chemical bond to such ceramics. The use of the silane agent increases the bonding effectiveness to the filler more than to the matrix resin. However, the views on silane treatment to resin composite are contradictory, and several studies have recommended silanization of composite material surfaces, whereas, according to others, silanization does not improve bond strength. Moreover, the silanized material inevitably undergoes some degree of hydrolysis over time, so achieving long-term stability is problematic. Thus, in the results of the present study, the Silane group also showed decreased bond strength after water storage; after 6 months of water storage, the value of bond strength was equivalent to those of the Control group and the Primer group.

In the present study, not only SEM observation, but also EDS elemental analysis of the interface was performed to investigate the bonding mechanism between the resin block and cement. On EDS analysis, mapping of various elements was performed, and C and Si were important to confirm the interface in detail (Fig. 5). On SEM observation, a bonding interface with a clear boundary was formed by control and silane treatment, whereas a thick intrusion layer was observed at the interface in the group treated with MMA-containing primers (i.e., both the Primer group and the Silane+Primer group) (Fig. 6). In our previous study, we confirmed that a thick intrusion layer of about 5 µm was formed at the MMA-containing primer-treated interface. In the present study, details of the intrusion layer were observed under EDS, and MMA-containing primer penetration with a depth of about 1–2 µm on the surface of the resin block was seen (Fig. 5b). The carbon distribution ratio was higher than that of the matrix resin part of cement, and it was considered that the carbon of the permeation part was likely to be derived from the MMA-containing primer. Since the carbon components of the matrix resin and the MMA-containing primer are similar, it is not possible to draw a clear line, but it is highly possible that the MMA-containing primer has penetrated into the microcracks formed on the block surface. It has been reported that alumina blasting causes microcracks at a depth of about 1 to 2 µm on the block surface. Therefore, it was considered that the MMA-containing primer penetrated into the microcracks generated by alumina blasting treatment. From the above results, in the Silane+Primer group, the chemical and mechanical adhesiveness was improved by the penetration of the MMA-containing primer into the deeper layers in the microcracks, so that it is thought that the bond strength was higher than that of the Silane group and the Primer group.

Differences between the Primer group and the
Silane+Primer group were found on CA measurement and EDS liner analysis of C and Si. The CA measurement may be affected by the properties of the liquid, such as the difference in hydrophilicity and hydrophobicity. Therefore, in this experiment, to evaluate the effect on the MMA-containing primer, the measurement was performed using the same primer. The MMA-containing primer used in this experiment has high viscosity. Therefore, in this experiment, there was no group with MMA-containing primer treatment before silane treatment as in previous studies; when multiple treatment agents are applied, silane treatment is performed first\textsuperscript{16,26,33}. The CA measurement using the resin primer showed that silane treatment decreased the CA. This result directly showed that the Silane+Primer group had an enhanced surface in terms of resin primer infiltration to the block adhesion surface. Considering the results of this experiment, the wettability of the block surface is improved by applying the silane coupling agent, and the MMA-containing primer penetrates deeply into the microcracks generated by the alumina blasting treatment. It is considered that a high mechanical fitting force could be obtained at the interface by curing the MMA-containing primer. This is a reason for the improved bond strength of the Silane+Primer group and the difference on fracture analysis. Moreover, the elemental liner analysis showed the difference between the Primer group and the Silane+Primer group in terms of the width of the carbon-dense layer.

This study clarified the advantages of the MMA-containing primer treatment and combined use of the silane treatment/MMA-containing primer treatment for resin blocks, including long-term adhesiveness. The limitation of this study is that only one kind of resin primer, silane agent, and one kind of resin block. In theory, the components of the resin primer and silane agent are simple; thus, similar results would be expected even when using other materials. However, a large difference can be detected in resin blocks, and then further study may be required using another resin block. Currently, in clinical practice, the demand for CAD/CAM resin crowns made from resin blocks is increasing. Improving the adhesiveness to the resin block not only improves the reliability of the CAD/CAM resin crown, but also makes it possible to expand the application to laminated veneers and cantilever bridges. In addition, it is possible that unprecedented dental treatment will be realized by developing new resin blocks and processing technologies.

ACKNOWLEDGMENTS

This work was supported by JSPS KAKENHI Grant Numbers 17H04389. The authors would like to thank associate professor Satoshi YAMAGUCHI, professor Satoshi IMAZATO (Department of Biomaterials Science, Osaka University Graduate School of Dentistry), associate professor Mutsumo ARANISHI, Ms. Nao EGUCHI (Center for Scientific Instrument Renovation and Manufacturing Support, Osaka University), and Mr. Yosuke MURAKAMI (The Institute of Scientific and Industrial Research, Osaka University) for providing us with helpful comments on this study. Kuraray Noritake Dental are acknowledged for supporting this work through the donation of materials. The authors would also like to thank FORTE Science Communications, for their English language editing services.

CONFLICTS OF INTEREST

The authors report no conflicts of interest.

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