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Effect of Surface Preparation on the Failure Load of a Highly Filled Composite Bonded to the Polymer-Monomer Matrix of a Fiber-Reinforced Composite

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Abstract

Purpose: The purpose of the present study was to evaluate the effect of surface preparation on the maximum fracture load value of a highly filled composite bonded to the polymer-monomer matrix of a fiber-reinforced composite.
Materials and Methods: A polymer-monomer matrix was made by mixing urethane dimethacrylate and triethyleneglycol dimethacrylate at a ratio of 1:1 with camphorquinone and 2-dimethylaminoethyl methacrylate as a light initiator. The matrix was then polymerized in a disk-shaped silicone mold with a light-polymerizing unit. The flat surfaces of the polymer-monomer matrix disk were prepared in one of the following ways: (1) without preparation; (2) application of silane coupling agent; or (3) application of matrix liquid and prepolymerization. A highly filled composite material was applied and polymerized with a light-polymerizing unit. Additional test specimens made entirely of the polymer-monomer matrix were fabricated as references; the disk and cylinder were fabricated in one piece using a mold specially made for the present study (group 4). Half the specimens were thermocycled up to 10,000 times in water with a 1-minute dwell time at each temperature (5°C and 55°C). The maximum fracture load values were determined using a universal testing machine ($n = 10$).
Results: The maximum fracture loads for group 3 were significantly enhanced both before and after thermocycling, whereas the maximum fracture loads of group 2 were significantly enhanced before thermocycling ($p < 0.05$); however, the failure loads decreased for all groups after thermocycling ($p < 0.05$). All the specimens in groups 1 and 2 debonded during thermocycling. The failure load of group 3 was significantly lower than that of group 4 both before and after thermocycling ($p < 0.05$).
Conclusion: Within the limitations of the current in vitro study, the application and prepolymerization of a mixed dimethacrylate resin liquid prior to the application of a highly filled composite was an effective surface preparation for the polymer-monomer matrix of a fiber-reinforced composite; however, the bond durability may be insufficient.

Recently, polymer materials such as highly filled composites[1,2] or fiber-reinforced composites[3-5] have been used in dental practice. In removable prosthodontics, procedures have been developed for replacing the missing abutment teeth of a removable partial denture using a composite resin coping with a magnetic attachment keeper,[6] or for fabricating a direct root coping with a keeper by attaching a coping portion made of composite resin core material to a fiber-reinforced dowel.[7] These new modalities may represent new, viable treatment options; however, little information exists regarding how to connect or add composite to composite.[8,9] In the present study, it was assumed that a highly filled composite does not generally bond strongly with a fiber-reinforced composite and that there are suitable surface preparations to facilitate the bonding of these two materials.

As the first step toward the improvement of such bonding, this preliminary study evaluated the effect of the surface preparation on the maximum fracture load of a highly filled composite bonded to a polymer-monomer matrix used as the matrix of a fiber-reinforced composite for the fabrication of root copings, dowel and cores, or other prostheses.

Materials and methods

Urethane dimethacrylate (UDMA, Negami Chemical Industrial Co. Ltd., Ishikawa, Japan) and triethyleneglycol dimethacrylate (TEGDMA, NK-Ester, Shin-Nakamura Chemical Co. Ltd., Wakayama, Japan) were mixed at a ratio of 1:1. As a light initiator (0.7 wt%), camphorquinone (CQ, Tokyo Kasei Co. Ltd., Tokyo, Japan) and 2-dimethylaminoethyl methacrylate (DEAM, Tokyo Kasei Co. Ltd.) were used at a ratio of 1:2.[10]

The mixed dimethacrylate resin liquid was poured into a disk-shaped mold (10.0-mm diameter, 2.5-mm thick) made of a silicone material. The disk specimens were then initially polymerized in the silicone molds for 1 minute with a light-polymerizing unit (Visio Alfa, 3M ESPE, Seefeld, Germany) and finally polymerized with another light-polymerizing unit (UniXS II, Heraeus Kulzer, Wehrheim, Germany) for 3 minutes. After final polymerization, each disk was removed from the mold and embedded in an autopolymerizing resin material with an acryl ring. The surfaces of the specimens were abraded under running water with 400-grit silicon carbide paper.

The 60 abraded specimens were divided into three groups according to the type of surface preparation given: (1) without preparation, (2) application of silane coupling agent (Clearfil Ceramic Primer, Kuraray Medical Inc., Tokyo, Japan) without drying, and (3) application of the mixed matrix liquid and prepolymerization for 6 seconds with Visio Alfa. Sticky tape with a hole (6-mm diameter) and a Teflon ring (5.0-mm inner diameter, 6.0-mm outer diameter) were placed on the surface to define the bonding area on each specimen. A highly filled composite (Estenia C&B E1, Kuraray Medical Inc.) was applied inside the Teflon ring and then polymerized with UniXS II for 3 minutes. After the polymerization process was complete, the sticky tape and the Teflon ring were removed as gently as possible. Twenty additional test specimens made entirely with the polymer-monomer matrix were fabricated as references as a one-piece disk and cylinder using a silicone mold specially made for the present experiment (group 4) (Fig 1). Half the specimens were placed in a thermocycling apparatus (Thermal Shock Tester TTS 1, Thomas Kagaku Co., Ltd., Tokyo, Japan) and cycled for 10,000 cycles in clean water between 5°C and 55°C with a dwell time of 1 minute at each temperature. A compressive load was applied to each specimen in a shear testing apparatus, and the maximum fracture loads were determined using a universal testing machine (Autograph AGS-J, Shimadzu Corp., Kyoto, Japan) at a crosshead speed of 1.0 mm/min (Fig 2). Ten specimens were tested for each experimental group, and the means and standard deviations (SD) for the maximum fracture loads were calculated and statistically analyzed with two-way ANOVA. The variables were surface preparation and thermocycling. After two-way ANOVA, the Bonferroni correction was performed when appropriate at the 0.05 level of significance.

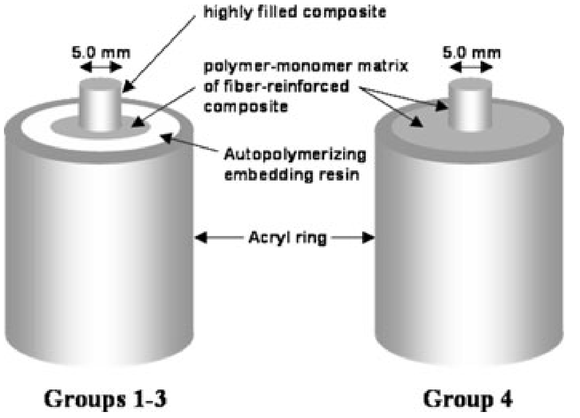

Figure 1 Specimen with highly filled composite bonded to polymer-monomer matrix (groups 1–3) and specimen made entirely from polymer-monomer matrix (group 4).

After testing, the interfaces of all specimens where failure occurred both before and after thermocycling were observed using an optical microscope (Nikon 92052, Tokyo, Japan) at 30× magnification. Failure was evaluated as A (adhesive failure at the matrix highly filled composite interface), C (cohesive failure within the polymer-monomer matrix without interface separation), or M (mixture of cohesive failure and adhesive failure).

Results

Two-way ANOVA and the Bonferroni correction revealed significant differences in the maximum fracture load value due to the variables of surface preparation and thermocycling ($p < 0.05$), whereas there were no significant differences in their interaction ($p > 0.05$). The maximum fracture loads of both groups 2 and 3 were significantly enhanced before thermocycling compared to group 1 ($p < 0.05$); however, the failure loads in all groups decreased after thermocycling ($p < 0.05$). All specimens in groups 1 and 2 debonded without exception during thermocycling. The failure load of group 3 was significantly lower than that of group 4 both before and after thermocycling ($p < 0.05$). The average maximum fracture loads, standard deviations, and statistical categories are summarized in Table 1.

The failure modes of all the specimens are presented in Table 2. In group 3, all specimens except one underwent cohesive failure in the 0 thermocycle condition, whereas seven specimens underwent mixed failure after thermocycling. In group 2, all specimens except one underwent mixed failure, whereas adhesive failure occurred in all the specimens after thermocycling. Adhesive failure, however, occurred in almost all the specimens in group 1 from the beginning of thermocycling.

Discussion

The combination of a highly filled composite and a fiber-reinforced composite produced excellent mechanical properties in an in vitro study.[11] In contrast to that particular report, other studies have indicated clinical failures of combinations of these

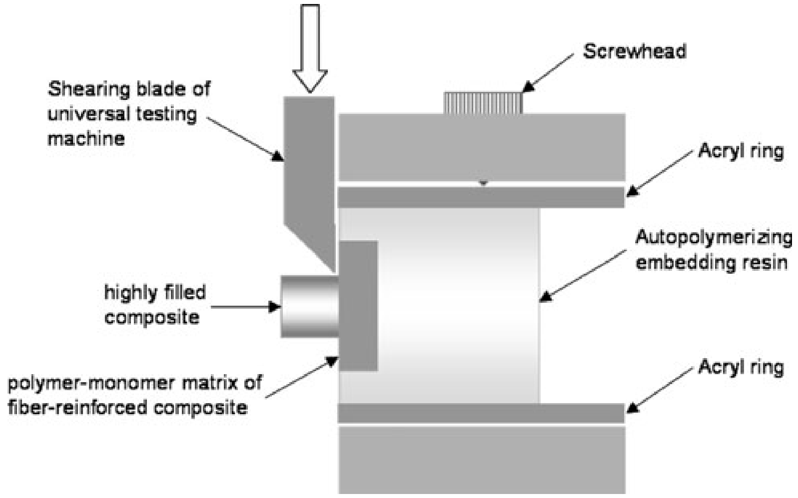

Figure 2 Specimen on shear testing apparatus in a universal testing machine.

two materials.[12] Sound bonding of highly filled composites to fiber-reinforced composites should be a matter of great importance, and an effective surface preparation for fiber-reinforced composite is required. In an effort to improve the bonding, the present study evaluated the effect of surface preparation on the maximum fracture load of a highly filled composite bonded to a polymer-monomer matrix of fiber-reinforced composite. The ultimate goal of this series of studies was to gain sufficiently high-bond strength between a highly filled composite and a fiber-reinforced composite so that they perform like one integrated bulk.

The failure load unit used in this study was N instead of MPa, which is a bond strength unit, since the gross area of the failure had not been defined when the cohesive failure occurred inside the substrate of the interface between the dimethacrylate resin material without the matrix and the highly filled composite. In such cases, the unit of strength is no longer available. Particularly in the typical case of specimen A in Figure 3, the failure occurred not only in the polymer-monomer matrix or the matrix highly filled composite interface but extended to the embedding material.

When denture base resins were repaired with a light-polymerized or autopolymerizing resin, the former exhibited a lower repair strength and toughness than the latter.[13] According to the manufacturer's product information, the resin component accounts for only 8% (18 vol%) of the highly filled composite used in this study. Considering these facts, it was assumed that the surface of the polymer-monomer matrix required some preparation to bond with the light-polymerized highly filled composite. The results showed that the application of the mixed matrix liquid and prepolymerization significantly improved the performance of the polymer-monomer matrix adhesives. This finding was confirmed by the failure mode; almost all of the failures in group 3 in the 0 thermocycle condition were cohesive (Table 2). On the other hand, the failure load value for the group without preparation was exceedingly low. Consequently, the presence of an adequate amount of residual monomer on the surface of the polymer-monomer matrix is considered to be indispensable for bonding to another light-polymerized resin material. A prepolymerized matrix may work like a bonding agent in restorative dentistry;[14] however, no cohesive failure occurred in any of the groups after thermocycling, indicating that the bond durability may be insufficient even when the mixed matrix liquid and prepolymerization are applied.

A silane coupling agent has been used in composite materials to coat a filler that chemically binds them to a resin matrix.[9] One study reported that the use of a bonding agent after the

Table 1 Maximum fracture loads (N) before and after thermocycling

Thermocycles	0 Cycles			10, 000 Cycles		
Group	Mean	SD	Significance	Mean	SD	Significance
1	104.3	45.7	a	0	0	a
2	365.4	48.9	b	0	0	a
3	421.1	53.1	c	268.2	87.5	b
4	674.3	39.3	d	335.3	59.0	c

Groups: 1, without preparation; 2, application of silane coupling agent; 3, application of mixed matrix liquid and prepolymerization; 4, entire polymer-monomer matrix.

SD = standard deviation. Identical letters indicate that the values are not statistically different ($p > 0.05$).

Table 2 Failure mode

Thermocycles Group	0 Cycles	10, 000 Cycles
1	M A A A A A A A A A	A A A A A A A A A A
2	C M M M M M M M M M	A A A A A A A A A A
3	C C C C C C C C C A	M M M M M M M A A A

Groups: 1, without preparation; 2, application of silane coupling agent; 3, application of mixed matrix liquid and prepolymerization.

A, Adhesive failure at the matrix-highly filled composite interface; C, Cohesive failure within the substrate polymer-monomer matrix without interface separation; M, Mixture of cohesive failure and adhesive failure. Each letter code corresponds to a separate specimen.

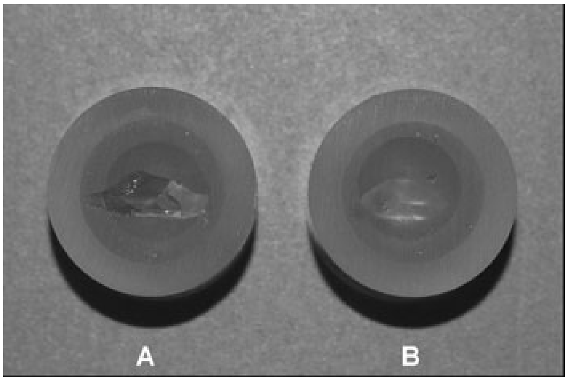

Figure 3 Cohesive failure within polymer-monomer matrix without matrix-highly filled composite interface separation (A) and mixture of cohesive failure and adhesive failure (B).

application of a silane coupling agent ensured adequate repair bonding between the layers of the composite material.[15] However, the crucial difference in the present study was that the surfaces to be bonded were not composed of composite material; they were made instead of a polymerized dimethacrylate resin with no filler. Unpolymerized highly filled composite resin paste material should be connected during the laboratory procedure to the polymerized fiber-reinforced composite on the cast to complete the prosthesis. Therefore, in the present protocol, the silane coupling agent must react with the filler particles in the highly filled composite, which is polymerized after coming into contact with the polymerized polymer-monomer matrix. Accordingly, in group 2, the surfaces of the polymer-monomer matrix coated with the silane coupling agent were kept wet, and then the highly filled composite paste was added and polymerized; however, the effect of the silane coupling agent was significantly inferior to that of the mixed matrix liquid and prepolymerization, although the highly filled composite used in this study was totally filled [92 wt% (82 vol%)]. Of particular note was that the bond strength of group 2 was exceedingly low after thermocycling, as shown in Table 1. Therefore, sufficient bond durability cannot be gained by the application of silane coupling agent. Overall, the failure modes obviously supported the results of the maximum fracture load values.

Further studies associated with this combined use of a highly filled composite and a fiber-reinforced composite may be necessary. The ultimate goal of the present study was to gain sufficiently high bond strength between the two materials so that they performed like one integrated bulk. A polymer-monomer matrix substrate without glass fibers was used as a preliminary step. The fiber-reinforced composite, which is composed of this matrix with glass fiber, will be evaluated for clinical use in the next study.

Conclusions

Within the limitations of the present study, the application of a mixed dimethacrylate resin liquid and prepolymerization is an effective surface preparation for the polymer-monomer matrix of a fiber-reinforced composite to be bonded to a highly filled composite; however, the bond durability may be insufficient.

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