Dental Materials Journal 2008; 27(5): 723−729

Original Paper

Effect of commercially available bonding agents impregnated with fibers on bending strength of hybrid resin

Sumio TSUSHIMA[1], Harunori GOMI[1], Akikazu SHINYA[1,2], Daiichirou YOKOYAMA[1], Pekka K. VALLITTU[2] and Akiyoshi SHINYA[1]

[1]*Department of Crown and Bridge, School of Life Dentistry at Tokyo The Nippon Dental University, 1-9-20 Fujimi, Chiyoda-ku, Tokyo 102-8159, Japan*

[2]*Department of Prosthetic Dentistry and Biomaterials Science, Institute of Dentistry, University of Turku, Lemminkäisenkatu 2, FI-20520 Turku, Finland*

Corresponding author, Akiyoshi SHINYA; E-mail: shinya-a@tky.ndu.ac.jp

To clarify the mechanical properties of fiber-reinforced hybrid resin bridges, this study evaluated the influence of various bonding agents ((Modeling Liquid (ML), DE Resin (DE), Bell Bond (BE), Mega Bond (MG), Durafil Bond (DU), Fluoro Bond (FB), Mac-Bond (MC), EG Bond (EG), Unifill Bond (UN), Single Bond (SN)) impregnated with fibers on bending strength. FB attained the highest bending strength of 570 MPa, whereas SN exhibited the lowest value of 224 MPa, which meant that the bending strength of FB was 2.5 times higher than that of SN. Results of this study suggested that the bending strength of fiber-reinforced hybrid resin was significantly affected by bonding agents impregnated with fibers. Therefore, selection of bonding agent for hybrid resin restoration requires careful consideration of product composition to ensure an optimal bonding agent-fiber combination, thereby imparting improved mechanical properties to the resultant dental restoration.

Key words: Fiber-reinforced material, Hybrid resin, Bonding agent

Received Feb 14, 2008: Accepted Apr 18, 2008

INTRODUCTION

Hybrid resin, originally introduced as esthetic restorative material for the posterior region, has been widely used for prosthodontic applications[1)]. When applying hybrid resin to a bridge, it is critical to reinforce the resin matrix with materials such as glass fibers. Studies so far revealed that the effectiveness of fiber reinforcement is dependent on many variables, including the form and diameter of fibers, quantity of fibers in the matrix, fiber placement and position, as well as the adhesion between fibers and matrix polymer[2-7)].

To reinforce hybrid resin with glass fibers, a bundle of fibers are to be immersed in a resin monomer (bonding agent in this study) and firmly bonded. Previous studies reported that fractures of fiber-reinforced hybrid resins were usually initiated at the interface between the fibers and resin matrix[8-10)]. It is therefore noteworthy that the bond strength between fibers and resin matrix differs significantly with bonding agents embedded with fibers, whereby the latter is thought to have a significant influence on reinforcing effect. Of particular importance too is the compatibility between the glass fibers and adhesive bonding agent when applying glass fibers to an inlay bridge and/or an adhesive bridge for intraoral direct repair[11,12)]. Therefore, the influence of bonding agent on bond strength is an important clinical guideline for the success of fiber-reinforced hybrid resin bridges and which demands careful evaluation.

To elucidate the mechanical properties of fiber-reinforced hybrid resin bridges, this study evaluated the influence of various commercially available bonding agents reinforced with fibers on bending strength. In addition, the fractured surfaces were observed by scanning electron microscopy (SEM).

MATERIALS AND METHODS

Materials used

Table 1 presents the manufacturers, lot numbers, compositions, and codes of bonding agents, restorative material, and glass fiber used in this study. Ten different kinds of commercially available bonding agents (ML, DE, BE, MG, DU, FB, MC, EG, UN, SN) were selected for evaluation. Each experiment was repeated six times, and thus a total of 60 experiments were carried out randomly.

Specimen preparation

Rectangular specimens of 3×4×40 mm dimensions were fabricated using a specially designed stainless steel slot located at the center of a mold. Bending test specimens were made by precutting glass fibers (ϕ11 μm) to a length of 40.0 mm. To remove constriction and lubricant, the 40.0-mm fibers were

rinsed with ethanol and purified water for 10 minutes each, air-dried for 24 hours, and then immersed in bonding agent for an hour. Excess bonding agent was removed with a wiper cloth (Kimwiper S-200, Crecia Corp., Tokyo, Japan).

Fibers which had been immersed in each bonding agent were placed perpendicularly to the base of the slot in stainless steel mold, and Estenia C&B (ES) to be filled on top. Both sides of the filled mold were pressed with glass slabs, and then light-polymerized with a light curing unit (Dentacolor XS, Kulzer, Hanau, Germany) for 90 seconds on each side for a total time of 180 seconds on both sides. To achieve complete polymerization, the specimens were transferred to a preheated heat/vacuum curing unit (KL-100, Kuraray Medical., Tokyo, Japan) and cured for 15 minutes at 110°C. For each piece of 3×4×40 mm ES test specimen prepared, the weight was 1.18 g while that of fiber was 0.19 g. In other words, fiber-weight ratio was 16.2 wt%. After polymerization, the marginal areas of specimens were polished with a #600 waterproof abrasive paper and stored in water at 37°C for 24 hours. Specimen preparation was done under Class 2 standard temperature and humidity conditions (23±2°C, 50±5%).

Table 1 Materials used in this study

Name	Code	Manufacturer	Lot Number	Main Components
Modelling Liquid	ML	Kraray Medical, Tokyo, Japan	00007A	MDP, UTMA, Cross-Linking Monomer, Photopolymerization catalyst
DE RESIN	DE	Bisco, Inc., Schaumburg, U.S.A	9.9E+09	UDMA, Bis-GMA, HEMA, DMPT Catalyst, Photopolymerization catalyst, Acetone
BELL BOND	BE	Kracie, Tokyo, Japan	A94	Phosphoric methacrylate, Bis-GMA, TEGDMA, Photopolymerization catalyst
MEGA BOND	MG	Kuraray Medical, Tokyo, Japan	0059AB	MDP, HEMA, MF, Photopolymerization catalyst
DURAFIL BOND	DU	Heraeus Kulzer, Hanau, Garmany	2.05E+08	Methacrylate solvent, Silicon dioxide, Benzoin methyl ether, Photopolymerization catalyst
FLUORO BOND	FB	Shofu, Kyoto, Japan	109963	UDMA, UIMA, Cross-Linking Monomer, Photopolymerization catalyst
MAC-BOND	MC	Tokuyama Dental Co., Tokyo, Japan	141	Bis-GMA, TEGDMA, HEMA, MAC-10, Water, Photopolymerization catalyst
EG BOND	EG	Sun Medical, Kyoto, Japan	TF3	4-META, Monomethacrylate, Dimethacrylate, Photopolymerization catalyst, Water
UniFil BOND	UN	GC, Tokyo, Japan	9906281	UDMA, TEGDMA, Photopolymerization catalyst, Water
Single Bond	SN	3M, St Paul Minnesota, USA	19991025	HEMA, Bis-GMA, functional methacrylate, Copolymer, Water, Ethanol, Photopolymerization catalyst
ESTENIA C&B	ES	Kuraray Medical, Tokyo, Japan		Monomer (Urethane methacrylatemonomer, Methacrylic acid monomer) Filler (Surface Treatment glass powder, Alumina system micro filler) Photopolymerization catalyst, colorant
Glass Fiber	GF	Nittobo, Tokyo, Japan		E-Glass (55wt%SiO_2, 15wt%Al_2O_3, 22wt%CaO)

MDP: 10-methacryloyloxydecyl dihydrogen phosphate
UTMA: Urethane tetra methacrylate
UDMA: Urethane dimethacrylate
DMPT: dimethyl-para-toluidine
TEGDMA: Triethyleneglycol dimethacrylate
HEMA: 2-hydroxyethylmethacrylate
MF: Mitogenic factor
MAC-10: 10-methacryloyloxy dideca methylene malonate
4-META: 4-methacryloxyethyl trimellitate anhydride
Bis-GMA: Bisphenol glycol mathacrylic acid

Bending test

Bending test was carried out in accordance with JIS bending test standard (JIS R1601) using a bending test device connected to a universal testing machine (Servopulser EHF-FD1, Shimadzu Co. Ltd., Kyoto, Japan). Each specimen was positioned such that GF was the tensile side. Load was then applied at a crosshead speed of 0.5 mm/min until fracture occurred. The ultimate load at fracture was recorded and used to calculate the bending strength in MPa using the following equation:

$$\delta = 3PL/2bh^2$$

where δ is the bending strength in MPa, P is the fracture load in N, L is the supporting width in mm, and b and h are the width and height of test specimen in mm respectively.

Statistical analysis

Bending strength data were evaluated statistically using one-way analysis of variance (ANOVA). Bonding agent was designated as factor A. After confirming that the X-R control limit of each value was equally dispersed, one-way ANOVA was performed on the data obtained. Where significant differences were noted, Tukey's multiple comparison test was carried out using a statistical software for data analysis.

SEM examination

After the bending test, fractured specimen surfaces were observed using a scanning electron microscope (S-4000, Hitachi Co. Ltd., Tokyo, Japan) at original magnification×500 and 5 kV accelerating voltage.

RESULTS

Bending strengths of different bonding agents reinforced with fibers

Table 2 presents the one-way ANOVA results obtained from the 10 commercially available bonding agents impregnated with fibers, whereby significant differences were found for factor A ($P<0.01$). Figure 1 shows the effects of bonding agents impregnated with fibers on bending strength. The mean values were 415, 522, 389, 441, 454, 570, 455, 323, 530, and 225 MPa for ML, DE, BE, MG, DU, FB, MC, EG, UN, and SN respectively. 95% confidence interval was Qi=39.4 MPa. One-way ANOVA demonstrated that FB attained the highest bending strength value of 570 MPa, whereas SN exhibited the lowest value

Table 2 One-way ANOAV results

factor	s. s	d. f	m. s	Fo
A: Bonding agent	569408	9	63267.6	2.07**
e	116285	50	2325.71	
T	685693	59		

**<0.01

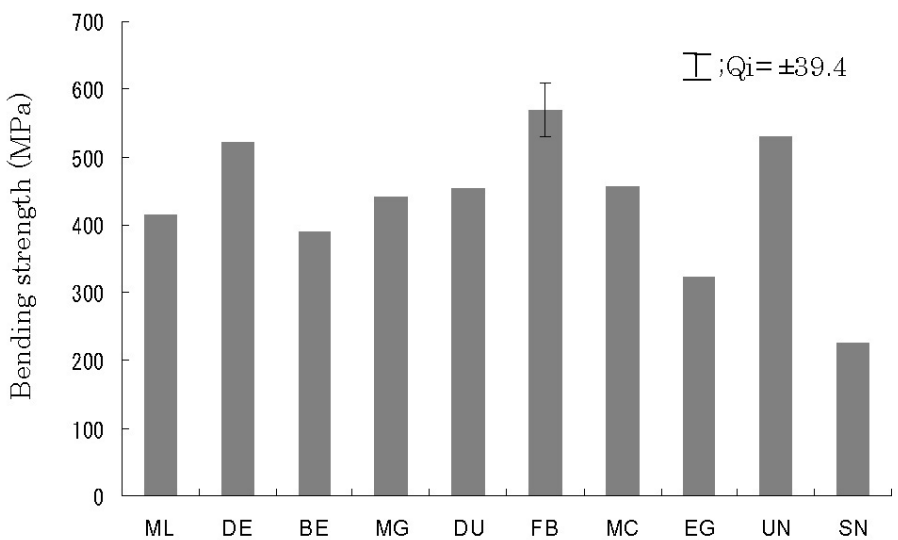
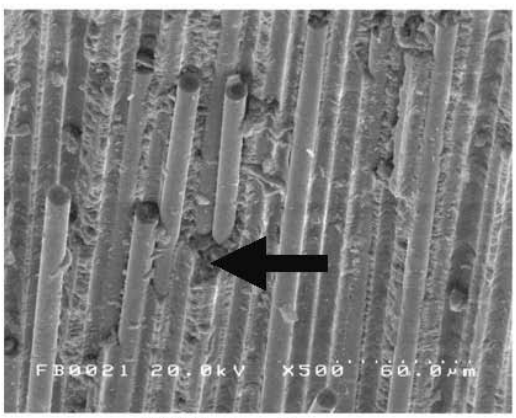


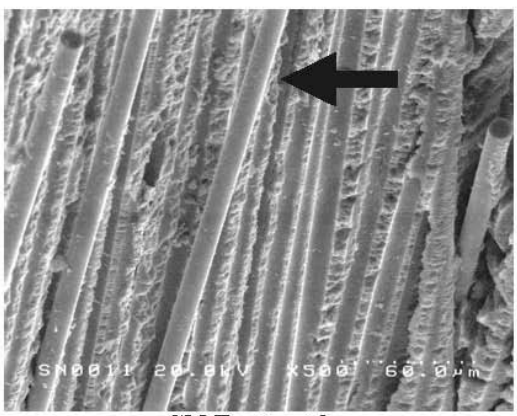
Fig. 1 Effects of bonding agents on bending strength.

Table 3 Tukey's multiple comparison test results

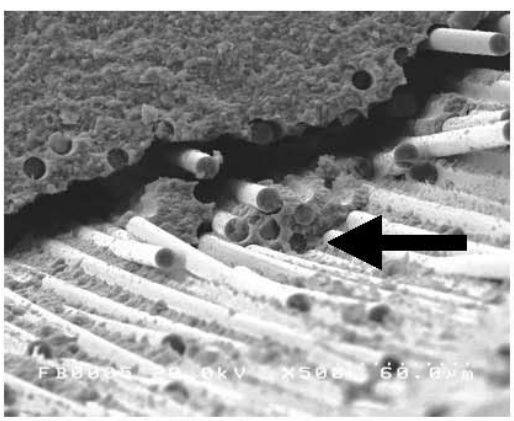
	ML	DE	BE	MG	DU	FB	MC	EG	UN
SN	＊＊	＊＊	＊＊	＊＊	＊＊	＊＊	＊＊	＊＊	＊＊
UN	＊＊		＊＊	＊＊	＊		＊	＊＊	
EG	＊＊	＊＊		＊＊	＊＊	＊＊	＊＊		
MC		＊				＊			
FB	＊＊		＊＊	＊＊	＊＊				
DU		＊							
MG		＊＊							
BE		＊＊							
DE	＊＊								



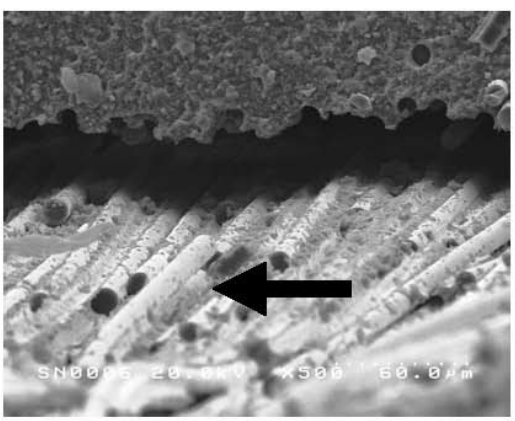
FB Resin side



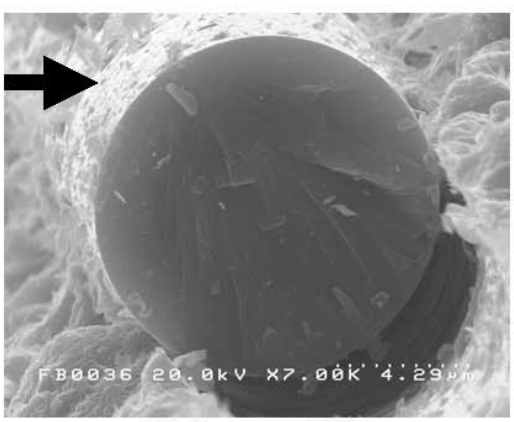
SN Resin side



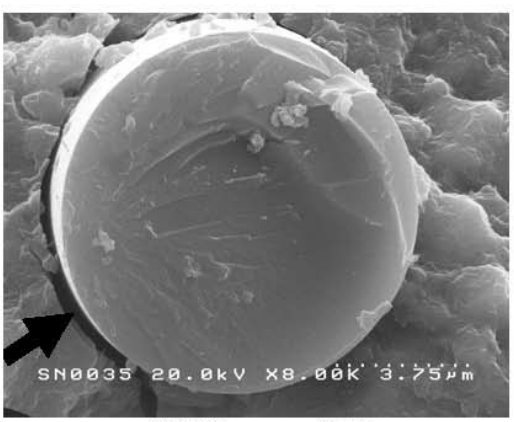
FB Fiber side



SN Fiber side



FB Closeup of Fiber



SN Closeup of Fiber

Fig. 2 SEM photographs of fractured surfaces pf glass fiber-reinforced hybrid composite.

of 224 MPa, which meant that the bending strength of FB was 2.5 times higher than that of SN. Table 3 lists the Tukey's multiple pairwise comparison results, whereby no significant differences were found between FB and DE, UN, but FB showed significant differences in other combinations. SN showed significant differences in all combinations. As for the hybrid resin ES, its bending strength was 168±21 MPa (n=6).

SEM observation of fractured surfaces

Figure 2 shows the SEM images of the fractured surfaces of FB (left side) and SN (right side) specimens, which attained the highest and the lowest mean bending strengths respectively. Topographic patterns observed on the resin and fiber sides are presented at the top and middle rows, while close-up views of the fibers are presented at the bottom row. For FB, failure mode was primarily mixed failure whereby the following topographic patterns were noted: concave surface caused by breakaway of fibers on the ES side, cutting plane of fibers, and slight hybrid resin attachment on the GF side. For SN, interfacial failure between hybrid resin and bonding agent was observed, featuring a concave surface caused by breakaway of fibers on the ES side as well as intact fiber surface on the GF side. DE and UN specimens demonstrated mixed failure, whereas ML, BE, MG, DU, MC, and EG specimens demonstrated interfacial failure. At the close-up view of fiber in FB specimen, fiber and resin were found to be bonded. In SN, however, detachment was shown at the interface of fiber and resin.

DISCUSSION

Test specimen preparation

There are two different techniques to fabricate fiber-reinforced composite bridges. One method is to light-cure fibers impregnated in a bonding agent and then build up hybrid resin on the polymerized surface, while the other method is to place hybrid resin on fibers which have been preimpregnated with a bonding agent. Whereas the former technique offers ease of handling, it runs the risk of air entrapment between hybrid resin and glass fibers, thus resulting in poor adherence. Though the latter offers enhanced adherence, it runs the risk of fiber movement during specimen preparation, as hybrid resin is placed on fibers which have been preimpregnated with a bonding agent and then pressed.

With an experiment using glass cloth and glass roving, Ellakwa *et al.*[3] reported that stronger fiber-reinforced plastic (FRP) frame was produced by pressing resin which had glass cloth and glass roving preimpregnated in matrix resin. Separately in other studies, test specimens were prepared — without polymerization of fibers — by placing hybrid resin on fibers which had been preimpregnated with a bonding agent. With this sequential placement method, no variations in data were observed. On this ground, the authors adopted the latter specimen preparation method.

Bonding agents are generally classified into two categories: direct intraoral application *versus* dental laboratory use. The former is to be applied to dentin cavity wall to increase surface free energy and to improve the wettability of the bonding agent on the dentin. This will consequently enhance the adhesion between tooth substrate and resin when filling the composite resin into the cavity. Its main composition consists of monomer, polymerization initiator, and solvent. Currently, in popular use are seventh-generation light-cure bonding systems that combine the etchant, primer, and adhesive into one bottle. As for bonding agents used in dental laboratories, they are to be applied to improve the wettability of the resin paste so as to facilitate the add-on procedure when fabricating facing crowns.

Effects of different bonding agents on bending strength

In addition to ML that was supplied with ES, nine different commercially available bonding systems were selected for this study. They could be divided into three groups depending on the adhesive system type: three-step adhesive (DU), self-etching adhesives (BE, MG, FB, MC, UN), and wet bonding systems (DE, EG, SN).

Bending test results revealed that the highest bending strength value of 570 MPa was achieved with FB, whereas the lowest value of 224 MPa with SN. Further, Tukey's multiple comparison test results (Table 3) showed that there were no significant differences among FB, UN, and DE specimens. Whereas the bending strength of Estenia stood at 168 MPa, the bending strengths of bonding agents impregnated with fibers were apparently improved, far exceeding this value. It is noteworthy that the three bonding agents which exhibited higher bending strengths (FB, UN, DE) contained urethane dimethacrylate monomer (UDMA), the same monomer as in the case of ES, thereby offering good affinity between ES and these bonding agents[8]. This was probably why tenacious bonding was achieved between ES and the glass fibers embedded in these bonding agents.

Upon examination of the fractured surfaces, FB, DE, and UN specimens — which obtained higher bending strengths — exhibited mixed failure, whereby ruptured fibers and slight hybrid resin attachment on fibers were noted. As for ML, BE, MG, DU, MC, EG, and SN specimens which achieved relatively lower bending strengths, interfacial failure

was exhibited between hybrid resin and glass fibers. The higher bending strengths of FB, DE, and UN specimens might be explained by the enhanced bonding between glass fibers embedded in the bonding agent and the hybrid resin. Consequently, the tenacious bonding between them led to an optimal reinforcing effect to resist increasing bending stress as load was applied. In addition, we speculated that the fractures, which were initiated at the fibers and cutting them off, propagated gradually through the entire specimens. In other words, this result showed that achieving enhanced bond strength for the bonding agent was critical to optimizing reinforcing effect[8].

In the present study, the inorganic filler contents of bonding agents were measured with ignition residue method. Bonding agent was incinerated at 575°C for an hour, and then the residual substances were measured. It was found that the inorganic filler contents were present at a level of 45 wt% on total composition for DU, 17 wt% for FB, 10 wt% for MG, 4 wt% for BE, and 2 wt% for UN; the remaining five bonding agents were not loaded with inorganic fillers. In other words, this finding suggested that higher bending strength was obtained by loading bonding agent with inorganic filler particles.

At this juncture, two reasons are proffered on why bending strength varied significantly with bonding agents. The first reason pertained to the adhesion between bonding agent and ES, and the other arose from the strength of fibers embedded in the bonding agent. Good adhesion was achieved when both the bonding agent and ES contained the same monomer; nonetheless, it was also necessary to increase the strength of the bonding agent itself[13-18].

Clinical application

Fiber reinforcement is effective only if the fiber, bonding agent, and hybrid resin each played its role and served its function fully. On the role and function of fibers, they should possess and exhibit an excellent stress-bearing capacity along the orientation of fibers, thereby bearing any increased stress generated. Fibers are to be firmly protected by the bonding agent[19-22]. Hence, for the bonding agent, it should not only provide adhesion between the fibers, but should also facilitate stress transfer from the resin matrix to the fibers. Therefore, future studies should be undertaken to further consider these two matters: how to increase adhesion between bonding agent and hybrid resin, and how to improve the strength of fibers embedded in bonding agent.

CONCLUSIONS

To clarify the mechanical properties of fiber-reinforced hybrid resin bridges, this study evaluated the influence of various bonding agents ((Modeling Liquid (ML), D/E Resin (DE), Bell Bond (BE), Mega Bond (MG), Durafil Bond (DU), Fluoro Bond (FB), Mac-Bond (MC), EG Bond (EG), Unifill Bond (UN), Single Bond (SN)) impregnated with fibers on bending strength. Besides, fractured surfaces were examined using SEM. Within the limitations of this *in vitro* study, the following conclusions were drawn:

(1) On the bending strength of fiber-reinforced hybrid resin, Fluoro Bond (FB) achieved the highest at 570 MPa, whereas Single Bond (SN) ranked the lowest at 224 MPa.

(2) SEM observation of fractured surfaces showed that FB, DE, and UN specimens exhibited mixed failure, featuring ruptured fibers and slight hybrid resin attachment on fibers. As for SN, ML, BE, MG, DU, MC, and EG specimens, they exhibited interfacial failure between hybrid resin and fibers.

Results of the present study showed that the bending strength of fiber-reinforced hybrid resin was significantly affected by bonding agents impregnated with fibers. Therefore, selection of bonding agent warrants careful consideration in view of its impact on the restoration's resultant mechanical properties.

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