Back to the multi-step adhesive system: A next-generation two-step system with hydrophobic bonding agent improves bonding effectiveness

Azusa YAMANAKA¹, Atsushi MINE¹, Mariko MATSUMOTO^{2,3}, Ryosuke HAGINO¹, Masahiro YUMITATE¹, Shintaro BAN¹, Masaya ISHIDA¹, Jiro MIURA⁴, Bart VAN MEERBEEK³ and Hirofumi YATANI¹

¹ Department of Fixed Prosthodontics, Osaka University Graduate School of Dentistry, 1-8 Yamadaoka, Suita-shi, Osaka 565-0871, Japan

² Department of Restorative Dentistry, Hokkaido University Graduate School of Dental Medicine, Kita 13, Nishi 7, Kita-ku, Sapporo-shi, Hokkaido 060-8586, Japan

³ KU Leuven (University of Leuven), Department of Oral Health Sciences, BIOMAT & UZ Leuven (University Hospitals Leuven), Dentistry, Kapucijnenvoer 7, box 7001, BE-3000 Leuven, Belgium

⁴ Division for Interdisciplinary Dentistry, Osaka University Dental Hospital, 1-8 Yamadaoka, Suita-shi, Osaka 565-0871, Japan
Corresponding author, Atsushi MINE; E-mail: mine@dent.osaka-u.ac.jp

This study evaluated the bonding effectiveness of a newly developed two-step hydrophobic bonding material. Three groups using different bonding systems were compared: BZF group, using the new bonding system (BZF-29; GC, Tokyo, Japan); GPB group, using a one-step bonding system (G-Premio Bond; GC); and SE2 group, using a two-step bonding system (CLEARFIL SE Bond 2; Kuraray Noritake Dental, Tokyo, Japan). Microtensile bond strength (μ TBS) was measured after storage in water for 24 h, 3 months and 6 months ($n=25$ /group). Fracture surfaces were observed under scanning electron microscopy. The BZF group showed significantly higher μ TBS than the other groups ($p<0.001$). Dominant failure patterns were cohesive failure for the BZF group (48–84%), mixed failure for the SE2 group (48–60%) and interface failure between adhesive and resin composite for the GPB group (48–52%). The hydrophobicity of the BZF-29 bonding system improves the long-term bonding effectiveness between adhesive and resin composite.

Keywords: Dental bonding, Dentin-bonding agents, Microscopy, Electron, Scanning, Hydroxyethyl methacrylate, Hydrophobic and hydrophilic interactions

INTRODUCTION

Dental restoration has been revolutionized by the development of resin composite¹. This treatment relies on the bonding effectiveness of adhesive materials including resin composites, which do not require the removal of sound dental structures for mechanical retention. Resin composite thus not only satisfies the esthetic demands of patients, but also is less invasive and reduces pulp irritation². However, deterioration of the adhesive interface over time is problem, and is caused by hydrolysis derived from dentin and/or adhesive. Clinically, this phenomenon is associated with the development of marginal leakage, which eventually leads to marginal discoloration and secondary caries^{3–5}. Therefore, bond strength and durability of the bond to dental hard tissue must be considered. Bonding to enamel is highly reliable, but bonding to dentin has been considered more difficult. Unlike enamel, dentin contains relatively large amounts of water, due to the connection to pulp tissue *via* a large number of fluid-filled tubules, moistening exposed dentinal surfaces^{6,7}. This fluid can cause hydrolysis reactions. Bonding to dentin is thus more difficult than bonding to enamel. Furthermore, since dentin is structurally hydrophilic, the adhesive is made more hydrophilic in order to adhere to the resin composite, which is hydrophobic, and that also can cause

hydrolytic deterioration of the adhesive itself.

Dentin adhesion currently implies the use of one of two approaches: the etch-and-rinse system, or the self-etch bonding system. Self-etch bonding systems predominate for bonding to dentin⁸. Self-etch bonding systems use either a one- or a two-step bonding system. One-step bonding systems involve a collection of all steps, and have recently become popular due to the simplicity of application. Basically, the one-step bonding system is more hydrophilic than the two-step bonding system⁹. However, these properties can lead to a loss of bonding effectiveness and stability. In fact, one-step bonding systems have been reported to offer inferior results to two-step bonding systems in terms of bonding effectiveness and durability^{10,11}. Furthermore, limits exist to the ability to combine the highly hydrophilic substance required for dentin adhesion and hydrophobic substances required for resin composite adhesion in a single step. Hence, two-step bonding systems are considered more practical than one-step systems⁵. As previously stated, the hydrophilicity and hydrolytic stability of adhesives are generally antagonistic, so the bond strength of adhesives over time is undermined by increasing concentrations of hydrophilic substances in adhesives. Two-step bonding systems offer better results than one-step bonding systems, but are still sensitive to hydrolytic degradation of ester bonds due to the hydrophilicity of the adhesive. In past studies, a method of coating a hydrophilic adhesive with more hydrophobic

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adhesive was proposed^{10,12)}.

BZF-29 (GC, Tokyo, Japan) is a newly developed two-step bonding system, in which the primer is designed based on technology used in G-Premio Bond (GC), a one-step bonding system, and hydrophobic bonding is achieved by removing 10-methacryloyloxydecyl dihydrogen phosphate (MDP) and hydroxyethyl methacrylate (HEMA), hydrophilic monomers, from the bonding resin. The hydrophobicity of BZF-29 bonding was evaluated based on water sorption and flexural strength measurements after storage in water, and was found to be higher than that for a gold-standard two-step system (CLEARFIL SE Bond 2, Kuraray Noritake Dental, Tokyo, Japan)¹³⁾. Hydrophilic monomers such as MDP and HEMA show high bonding strength of the adhesive with dentin as a primer, but concerns have been raised regarding the stability of adhesion to the resin composite, and a reduction in long-term durability due to deterioration resulting from water absorption^{14,15)}. The bond strength of adhesives may be improved if the adhesive resin covering the primer is more hydrophobic. Increasing the hydrophobicity of the adhesive may improve its long-term bond strength, but evidence confirming this has not yet been reported. The bond strength of the newly developed BZF-29 bonding system thus needs to be assessed in comparison to G-Premio Bond and the gold-standard two-step system as control groups.

The purpose of this study was to investigate the bonding effectiveness of the newly developed two-step bonding system after storage in water for 24 h, 3 months and 6 months. The null hypothesis to be tested was that the bonding effectiveness of the new two-step adhesive containing hydrophobic bonding would not be different from those of other adhesives.

MATERIALS AND METHODS

Tooth preparation and bonding procedures

The methods for tooth preparation and bonding procedures are shown in Fig. 1. Fifteen extracted, non-carious human molars, collected with the informed consent of the patients under a protocol reviewed and approved by the institutional review board of Osaka University, were used in the present study (approval No.: H30-E51). These molars were cut at the height of the contour and exposed dentin, then polished with #600 silicon carbide paper. Samples were randomly divided into three groups depending on the bonding material used. Three bonding systems were employed in the present study. The first was the newly developed experimental two-step bonding system, BZF-29 (GC, [BZF group]). The other two were a one-step bonding system, G-Premio Bond (GC, [GPB group]) and a two-step bonding system, CLEARFIL SE Bond 2 (Kuraray Noritake Dental, [SE2 group]). Table 1 shows the chemical formulations and instructions from the respective manufacturers for the application of these three adhesives. A resin composite (CLEARFIL AP-X; Kuraray Noritake Dental) was built up in 2-mm-thick layers at one time, then light-cured for

Fig. 1 Preparation of micro-specimens.

(a) Caries-free molar is cut at the height of the contour and exposed dentin. (b) The tooth is polished with #400 and #600 silicone carbide papers. (c) The dentinal surface is air-dried, then adhesive is applied and light-cured. (d) Resin composite is built up in 2-mm-thick layers and light-cured. (e) Specimens are stored in distilled water at 37°C for 24 h. (f) Specimens are sectioned in beam shapes with a cross-sectional area of 1 mm² ($n=25$). (g) The remaining beam-shaped specimens are stored in distilled water at 37 °C. (h) Values of microtensile bond strength (μ TBS) are measured and fractured surfaces of specimens after μ TBS measurement are analyzed by scanning electron microscopy (SEM).

20 s from four directions (Mini LED3, maximum light intensity 2,200 mW/cm²; Satelec, Merignac, France). Specimens were stored in distilled water at 37 °C for 24 h, and then sectioned into beams with a cross-sectional area of 1 mm². Microtensile bond strength (μ TBS) was then measured immediately or after storage in distilled water at 37 °C for another 3 months or 6 months ($n=25$ /group for each time point).

Testing of μ TBS

Each beam was attached to a Ciucchi's jig with a cyanoacrylate adhesive (Model Repair II Blue; Dentsply-Sankin, Tokyo, Japan) and subjected to tensile force at a crosshead speed of 1 mm/min in a desktop testing apparatus (EZ test; Shimadzu, Kyoto, Japan) until failure. Fractured specimens were then carefully removed from the jig. The value of μ TBS was expressed in megapascals, dividing the applied force (N) at the time of fracture by the bonded area (mm²). Mean bond strength of the 25 beams derived from each group represented the μ TBS of that group, generating three values for the water storage periods per group. The resultant μ TBS values from each system and storage period were evaluated with the Kruskal-Wallis test ($n=25$). A p -value less than 0.05 was considered statistically significant.

All statistical analyses were performed using EZR (Saitama Medical Center, Jichi Medical University, Saitama, Japan), a graphical user interface for R (The R Foundation for Statistical Computing, Vienna, Austria). More precisely, EZR is a modified version of R Commander designed to add statistical functions frequently used in biostatistics¹⁶⁾.

Fracture mode analysis

After μ TBS testing, modes of failure in all specimens were observed using a field emission scanning electron microscope (TM3000; Hitachi, Tokyo, Japan) with an accelerating voltage of 10 kV. Specimens were visually categorized by the same person as showing cohesive, mixed or adhesive failures. Cohesive failure was defined as 90–100% of the failure occurring in the dentin, adhesive or resin composite. Adhesive failure was defined as 90–100% of the failure occurring between the dentin and adhesive or between the adhesive and resin composite. Mixed failure was defined as 10% or more adhesive failure and cohesive failure in the dentin, adhesive and/or resin composite. In addition, more detailed failure modes were confirmed in this study. Details of the categories are: CR, cohesive in resin composite; Ad (r/ad), adhesive failure at resin composite/adhesive interface; M (r, ad), mixed failure at resin composite and adhesive; CAd, cohesive failure in adhesive; Ad (ad/d), adhesive failure at adhesive/dentin interface; M (ad, d), mixed failure at adhesive and dentin; CD, cohesive in dentin; and M (r, ad, d), mixed failure at resin composite, adhesive and dentin.

RESULTS

Findings for μ TBS

The initial μ TBS (mean $\pm$ SD) values were 62.9 $\pm$ 20.1,

29.0 $\pm$ 12.6 and 50.9 $\pm$ 16.3 MPa, and the values after 6 months were 63.7 $\pm$ 27.3, 23.6 $\pm$ 5.0 and 47.0 $\pm$ 26.2 MPa, for BZF, GPB and SE2 respectively (Fig. 2). Results from Kruskal-Wallis testing demonstrated that, among the parameters “water storage period” ($p>0.45$, Fig. 3a) and “adhesive systems” ($p<0.001$, Fig. 3b), only “adhesive systems” had any significant effect. In addition, μ TBS values were significantly higher in the BZF group than in the GPB and SE2 groups ($p<0.001$), also they were significantly higher in the SE2 group than in the GPB group ($p<0.001$). No pre-test failures were encountered in this study.

Fracture modes

Fracture modes were affected by the adhesive systems

Fig. 2 Box plot of data for microtensile bond strength (μ TBS).

Effect of storage in water on mean μ TBS of adhesive systems. Error bar, SD; center line, median; bottom of box, lower quartile (25%); top of box, upper quartile (75%).

Table 1 Adhesives (manufacturer/lot number), composition, and manufacturer instructions

Adhesive (manufacturer/lot number)	Composition	Manufacturer instructions
BZF-29 (GC/ 1-Primer: 1907172, 2-Bond: 1907201G)	1-Primer: 10-MDP, 4-META, 10-methacryloyloxydecyl dihydrogen thiophosphate, dimethacrylates, water, acetone, photo initiators, silica filler 2-Bond: dimethacrylates, bis-GMA, photo initiators, silica filler	1. Apply 1-Primer and leave for 10 s. 2. Dry thoroughly with air under maximum air pressure for 5 s. 3. Apply 2-Bond. 4. Gentle air blowing to make the film uniform. 5. Light cure for 10 s.
G-Premio Bond (GC/ 1909131)	10-MDP, 4-META, 10-methacryloyloxydecyl dihydrogen thiophosphate, dimethacrylates, distilled water, acetone, photo initiators, silica filler	1. Apply the adhesive and leave for 10 s. 2. Dry thoroughly with air under maximum air pressure for 5 s. 3. Light cure for 10 s.
CLEARFIL SE Bond 2 (Kuraray Noritake Dental/ Primer: 2D0109, Bond: 2K0170)	Primer: 10-MDP, HEMA, hydrophilic aliphatic dimethacrylate, dl-CQ, water Bond: 10-MDP, bis-GMA, HEMA, dl-CQ, hydrophobic aliphatic dimethacrylate, initiators, accelerators, silanated colloidal silica	1. Apply the primer and leave for 20 s. 2. Gentle air blowing for >5 s. 3. Apply the bond. 4. Gentle air blowing to make the film uniform. 5. Light cure for 10 s.

Fig. 3 Statistical results. Error bar, SD; center line, median; bottom of box, lower quartile (25%); top of box, upper quartile (75%).

Fig. 4 Details of fracture modes. CR, cohesive failure in resin composite; Ad (r/ad), adhesive failure at resin composite/adhesive interface; M (r, ad), mixed failure at resin composite and adhesive; CA, cohesive failure in adhesive; M (ad, d), mixed failure at dentin and adhesive; Ad (ad/d), adhesive failure at dentin/adhesive interface; CD, cohesive failure in dentin; M (r, ad, d), mixed failure at resin composite, dentin and adhesive. Interface failure between the adhesive and resin composite is frequent in the GPB group, whereas cohesive failure in the resin composite or dentin is frequent in all preservation periods in the BZF and SE2 groups.

(Fig. 4). Regardless of the water storage period, for both the BZF and SE2 groups, the predominant failure mode was cohesive failure. In detail, cohesive failure in the resin composite or dentin was observed in 48–84% and 40% of samples in the BZF and SE2 groups, respectively. However, interface failure was observed in 48–52% of samples in the GPB group, and most interfacial failure occurred between adhesive and resin composite.

The fractured surface on the dentin and resin composite side after μTBS test was observed under scanning electron microscopy (SEM) (Fig. 5). Typically, interface failure between the adhesive and resin composite was observed in the GPB group. Even within the same category of “mixed failure”, the BZF group and the SE2 group showed a difference in crack propagation

Fig. 5 SEM images of fractured surface in each group. a–d) BZF group. Mixed failure at dentin, adhesive, and resin composite is observed. Typical crack propagations in the adhesive layer (Δ). e–h) GPB group. Majority of interface failures occur between the adhesive and resin composite. Filled white arrows ($\uparrow$) in g show voids. i–l) SE2 group. Mixed failure at dentin and adhesive is observed. Crack propagation of the adhesive layer differs from that of the BZF group (Δ). D, dentin; Ad, adhesive; RC, resin composite.

within the adhesive layer. Linear crack propagation was frequently observed in the BZF group. A characteristic image of crack propagation is depicted in Fig. 5c and 5d.

DISCUSSION

To improve adhesion to dentin, bonding effectiveness of the new two-step system containing hydrophobic bonding was investigated in comparison to existing one- and two-step systems. In the present study, the bonding effectiveness of the new two-step adhesive, BZF, was significantly higher than that in the GPB and SE2 groups ($p < 0.001$). The null hypothesis that the bonding effectiveness of the new two-step adhesive containing hydrophobic bonding would not be different from other adhesives was thus rejected. The μ TBS test was used to mechanically assess the strength of the resin-dentin interface complex. The μ TBS test is considered one of the most reliable methods to evaluate bonding capability *in vitro*. The μ TBS was first evaluated at 24 h, but as bond strength tests should be performed to evaluate degradation over time, long-term experiments more closely resembling clinical situations also need to be assessed^{3,15}. In the present study, no decrease in bond strength was observed in all adhesives even after leaving sectioned beams in water for 6 months, allowing water to be in direct contact with the interface. From this result, all adhesives tested in the present study can basically be considered to offer excellent stability. In addition, fracture mode analysis was performed after μ TBS. Although no significant changes in fracture mode were seen between storage periods for GPB and SE2, BZF interestingly showed sequential increases in cohesive failures, from 48% to 84%. A cohesive failure with breaks in dentin or resin composite implies that the interface between the dentin and adhesive is sufficiently strong.

The bonding resin in BZF contains no HEMA, a hydrophilic and water-miscible monomer widely used in dental materials. The hydrophilicity and hydrophobicity of the adhesive are important for bonding effectiveness and durability, because the adhesive needs to bond to hydrophilic dentin and hydrophobic resin composite. HEMA is usually used to incorporate complex compositions, including hydrophilic and hydrophobic monomers, solvents, and water, within the simplified bonding system on dentin^{17,18}. Van Landuyt *et al.* conducted a study using experimental adhesives with different concentrations of HEMA, and found that a small amount of HEMA (10%) improved the bond strength of one-step self-etch adhesives¹⁹. Nishitani *et al.* conducted a study examining dentin bond strength after 24 h and 6 months using adhesives with various concentrations of HEMA. They reported that higher adhesive hydrophilicity was effective for short-term bond strength, but may cause lower bond strength after 6 months¹⁵. Good wetting, diffusion, and penetration properties thus allow high bond strength of the adhesion to dentin, but concerns have remained regarding the

stability of adhesion to resin composite and a reduction in long-term durability due to deterioration resulting from water absorption¹⁵. Moreover, removing MDP, which is a functional monomer, is also effective for improving the hydrophobicity of the bonding resin in BZF. Since HEMA and MDP are removed, even if BZF is a two-step system similar to existing types such as SE2, the first and second steps are referred to as “1st bonding with priming” and “2nd bonding without priming”, respectively.

GPB is the adhesive in the HEMA-free one-step system. Basically, in HEMA-free systems, the concentrations of solvent and water are high, so evaporating the solvent is difficult, and the adhesive layer absorbs water, which is seen between the adhesive and resin composite (Fig. 5g)^{17,20}. In the present study, to clarify the characteristics of the adhesive used, details of fracture mode were analyzed under SEM. In the GPB group, interfacial failure between adhesive and resin composite was the dominant failure pattern, at 48% after 6 months. On the other hand, interface failure was not observed in the BZF group (0%) or SE2 group (0%) after 6 months. Basically, with one-step adhesive, the hydrophilic substance required for dentin adhesion and the hydrophobic substance required for resin composite adhesion need to be combined, placing a limit on increasing hydrophobicity. The bonding resin after using primer in a two-step system can be more hydrophobic than a one-step adhesive. From these results, in theory, none of the contemporary one-step adhesives can compete with a two-step system in terms of bonding effectiveness. In addition, bond strength reportedly increased when a one-step system as a primer and a bonding resin from a two-step bonding system was used²¹. This is because the mechanical properties of the adhesive layer are also increased by sealing the hydrophobic bonding resin on one-step adhesive, which is hydrophilic for dentin bonding. A strong correlation was found between resin-dentin bond strength and the mechanical properties of adhesives²². BZF has been reported to show higher mechanical strength than SE2¹³, and the linear crack propagation frequently observed in the BZF group may be related to this. Therefore, two-step systems are superior to one-step systems in terms of both bonding effectiveness and durability.

SE2 is considered a gold-standard two-step system because of its good *in vitro* and clinical performance^{22,23}. On the other hand, bond strength reportedly decreased after storage for 1 year^{23,24}. The decrease in bond strength of hydrophilic adhesive after 1 year of storage in water is related to the water absorption of HEMA, which reduces the mechanical strength of the resin²⁵. In the present study, bond strength was tested and interfacial appearance under SEM was observed after 24 h, 3 months and 6 months, but more long-term experiments and clinical studies are needed in the future. Observation of the bonding interface under transmission electron microscopy (TEM) to examine the interface between adhesive and dentin in detail is also necessary. In addition, hydrophobicity of the adhesive should be

evaluated, along with the degree of polymerization of the adhesive. BZF has different primer and bonding resin compositions compared to the gold-standard adhesive SE2. However, the present study revealed that the newly developed adhesive BZF offers excellent bonding potential due to the hydrophobic bonding resin.

CONCLUSION

The newly developed, two-step hydrophobic bonding material BZF showed higher bonding effectiveness than the gold-standard SE2 two-step system and the GPB one-step system. BZF improved bonding between adhesive and resin composite.

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