A comparison of dentin bond strength and degree of polymerization of bulk-fill and methacrylate-based flowable composites

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Abstract
Objective: This study aimed to evaluate the dentin bond strength and degree of polymerization of bulk-fill and methacrylate-based flowable composites.

Materials and Methods: Dentin bond strength of composites was evaluated using the microtensile bond strength (µTBS) test. Five different composites were applied on flat dentin surfaces with self-etch and etch-and-rinse adhesives. Twenty microspecimens with a 1 mm² cross-sectional area were prepared for each group (n = 20). The µTBS test was then applied. To measure the degree of polymerization of composites, eight cylindrical blocks were prepared (n = 8) from each composite. The surface microhardness of each composite was measured on the bottom and top surfaces, and a hardness ratio (HR) was calculated. Data were analyzed using one-way ANOVA and the Tukey’s honestly significant difference test (α = 0.05).

Results: According to the µTBS test results, the SDR Etch-and-Rinse group exhibited the highest dentin bonding strength and the Aelita self-etch group exhibited the lowest (P < 0.05). HR measurement showed that all composite groups had a sufficient degree of polymerization at a 2-mm thickness, while only the SDR and Xtra-Base groups had sufficient polymerization at a 4-mm thickness.

Conclusions: Bulk-fill composite demonstrates an even higher degree of polymerization of methacrylate-based flowable composite when applied in a 4-mm thickness.

Keywords: Bulk-fill flowable composite; degree of polymerization; dentin bond strength; hardness ratio; methacrylate-based flowable composite

INTRODUCTION

Flowable composites were developed to create a stress-reducing barrier during restoration with conventional resin composites. In order to reduce viscosity and for ease of application, the amount of filler used in flowable composites is 20%–25% lower compared to that of conventional methacrylate-based composites.[1] Due to this low filler content, methacrylate-based flowable composites have lower dentin bonding strength and greater polymerization shrinkage than methacrylate-based composites.[2,3] During the polymerization of composite resins, the amount of conversion of monomers to polymers is known as the “degree of polymerization.”[4] The degree of polymerization of composite resin affects its physical and mechanical properties. In case of insufficient polymerization, problems such as marginal leakage, staining, increased erosion, water absorption, and low mechanical resistance have been reported at restoration.[5]

The layering technique has been proposed to reduce the degree of polymerization of composite resins, but this method leads to loss of time in clinical practice.
Recent advances in composite technology for posterior restoration have been proposed as alternatives to the incremental layering technique. For this purpose, bulk-fill flowable composites have been developed which possess greater mechanical properties and polymerization kinetics than methacrylate-based flowable composites. Bulk-fill composites’ improved translucent structure and photoactive groups disposed within methacrylate resin give these composites better control of polymerization kinetics, and the composite can be employed using a bulk technique to a depth of 4 mm.

The dentin bond strength of flowable composites is an important factor in restoration success. To date, there has been insufficient investigation of the dentin bond strength of methacrylate-based flowable composites compared to bulk-fill flowable composites. This study compared the dentin bond strength and degree of polymerization of bulk-fill and methacrylate-based flowable composites. The following null hypotheses were evaluated: (1) There would be no differences in dentin bond strength between bulk-fill and methacrylate-based flowable composites and (2) bulk-fill flowable composites would exhibit a sufficient degree of polymerization at 4-mm thickness.

**MATERIALS AND METHODS**

Ethical approval (no: 29/2013) was obtained from the Ethics Committee, Faculty of Dentistry, Ataturk University. The chemical composition and manufacturer details of the materials used are listed in Table 1.

**Dentin bond strength test**

Fifty sound human molar teeth were used. Occlusal enamel was removed under water cooling. A standard smear layer was created on the dentin surfaces with 600-grit silicon carbide abrasive for 60 s under water. Then, teeth were divided into five groups, and each group was divided into two subgroups. Self-etch adhesive was applied to one of the each composite subgroup, while Etch-and-Rinse was applied to the other. Three bulk-fill flowable composites (Surefil SDR, Dentsply Konstanz, Germany; Xtra-Base®, Voco Cuxhaven, Germany; and Filtek™ Bulk Fill, 3M™ ESPE®, Seefeld, Germany) and two conventional methacrylate-based flowable composites (Elite™ Flow, Bisco Inc., Schaumburg, Illinois, USA, and GrandioSO Flow Voco Cuxhaven, Germany) were applied to both self-etch and etch-and-rinse groups. In the self-etch groups, Clearfil SE Bond (Kuraray Medical Inc., Japan) was applied to all dentin surfaces with a disposable applicator for 15 s and then cured for 10 s using an Elipar S10 LED curing unit (3M™ ESPE™ Dental Products, St. Paul, MN, USA) at an intensity setting of 1000 mW/cm².

In the etch-and-rinse groups, Scotchbond 37% orthophosphoric acid (3M™ ESPE®, Seefeld, Germany) was applied to all dentin surfaces for 15 s. The acidic surfaces were then washed for 10 s with water spray, air-dried for 5 s, and the acid was removed. Adper Single Bond 2 adhesive (3M™ ESPE™, Seefeld, Germany) was applied to all dentin surfaces using a disposable applicator and cured for 10 s. A 4-mm flowable composite was placed on the dentin surfaces to which adhesive was applied with mild air and cured for 40 s. Following 24-h storage in 37°C distilled water, specimens were placed on an L-shaped acrylic block, and 1 mm ± 0.2 mm² microspecimens were prepared using a low-speed diamond saw (ISOMET, Buehler, Evanston, IL, USA). Twenty microspecimens were prepared for each group. These microspecimens were placed on a digital microtensile bond strength (μTBS) test device (Microtensile Tester, Bisco, Inc., Schaumburg, IL, USA) with a cyanoacrylate adhesive (Zapit, Dental Ventures of America, Inc., Corona, CA, USA). μTBS testing was performed at a crosshead speed of 1 mm/min until the failure in the specimens. μTBS (MPa)

**Table 1: Materials, manufacturer, contents, filler, and batch numbers used in this study**

| Material                  | Manufacturer          | Type                | Matrix type                      | Filler type                           | Filler volume (%) | Batch number |
|---------------------------|-----------------------|---------------------|----------------------------------|---------------------------------------|-------------------|--------------|
| Surefil SDR               | Dentsply Konstanz, Germany | Bulk-fill composite | Polymerization modulator, UDMA, Bis-GMA, UDMA, TEGDMA | Ba-B-F-Al silicate glass, SiO₂, Inorganic filler | 44                | 131200155 |
| Xtra-Base                 | Voco Cuxhaven, Germany | Bulk-fill composite | Bis-GMA, UDMA, TEGDMA            | 58                                    | 1422161 |
| Filtek Bulk Fill          | 3M™ ESPE®, Seefeld, Germany | Bulk-fill composite | Bis-GMA, UDMA, Bis-EMA           | Zirconia and silica (0.01-3.5 µm)    | 42.5 | 4868A1 |
| Elite Flow                | Bisco Inc., Schaumburg, Illinois | Low-viscosity flowable composite | Bis-GMA, TEGDMA                   | Glass frit                          | 42               | 1404511 |
| GrandioSO Flow            | Voco Cuxhaven, Germany | Two-step self-etch adhesive | Methacrylate                     | Pirojenic silica, glass ceramic, Floride stabilizer | 81               | 1331164 |
| Adper Single Bond 2       | 3M™ ESPE®, Seefeld, Germany | Two-step self-etch adhesive | Methacrylate photoactivator, stabilizer | Nonspecific                        | N499596 |
| Clearfil SE Bond          | Kuraray Medical Inc., Tokyo, Japan | Two-step self-etch adhesive | MDP, HEMA, Bis-GMA, hydrophilic dimethacrylate, colloidal silica | MDPB, MDP, HEMA, hydrophilic dimethacrylate | Nonspecific    | 6K0040 |

UDMA: Urethane dimethacrylate, Bis-GMA: Bis-phenol A diglycidylmethacrylate, HEMA: 2-hydroxyethyl methacrylate, TEGDMA: Triethyleneglycodimethacrylate, MDP: 10-methacryloyloxymethyl dihydrogen phosphate
was calculated by dividing the failure load (N) by the cross-sectional area (1 mm²) of the test bar, and the data obtained were recorded.

**Degree of polymerization by Vickers microhardness**

In this study, the degree of polymerization of composites was evaluated by the method specified by Bouschlicher et al.[9] For hardness ratio (HR) measurement, two groups with 2 mm and 4 mm thickness were prepared from each composite, so a total of ten groups were prepared. Eight samples were prepared in each group (n = 8). Polytetrafluoroethylene cylindrical molds, 4 mm and 2 mm long with an internal diameter of 6 mm, were used to prepare the composite specimens. The mold was placed on a glass slide covered with a Mylar strip, and the composite was filled by syringing in bulk for each material. The specimens were then cured on the upper surface with an Elipar S10 LED curing unit (3M™ ESPE®, Seefeld, Germany) for 40 s, keeping the tip of the light-curing unit in contact with the specimens. Following 24-h storage in 37°C distilled water, microhardness measurements were performed on the top and bottom surfaces of each composite specimen using a Vickers Micro-hardness Tester (MVK-H1, Akashi Co., Tokyo, Japan) with a 200 g load over 10 s. HRs for all specimens were calculated using the formula: \( HR = \frac{V_{\text{bottom}}}{V_{\text{top}}} \). The specimens with a HR of 0.80 or above were considered sufficiently polymerized.

**Statistical analysis**

\( \mu \)TBS and HR values were analyzed using one-way ANOVA, the Tukey’s honestly significant difference test, and Student’s t-test (α = 0.05) on SPSS 20.0 software (SPSS Inc., Chicago, IL, USA).

**RESULTS**

\( \mu \)TBS data of flowable composites using self-etch and etch-and-rinse adhesives were compared using one-way ANOVA, and the mean values are shown in Table 2. Statistically significant differences were determined between the bond strength of dentin adhesive systems and flowable composites (\( P < 0.05 \)). The highest bond strength was observed in the SDR etch-and-rinse group (20.36 MPa) and the lowest average bond strength was observed in the Aelite self-etch (12.77 MPa) group. The SDR, Aura, and Aelite etch-and-rinse groups exhibited statistically significantly higher bonding strengths than self-etch groups (\( P < 0.05 \)). No significant differences were determined between the etch-and-rinse and self-etch groups in other composites (\( P > 0.05 \)).

Vickers microhardness measurement results are shown in Table 3. The highest top surface microhardness was observed in the Grandio groups and the lowest in the Aelite groups. In the 4-mm groups, statistically significant differences were observed between top surface microhardness and bottom surface microhardness (\( P < 0.05 \)), while in 2-mm groups, statistically significant differences were observed only in the Grandio and Aelite groups (\( P < 0.05 \)).

In the 2-mm groups, the highest HR was determined in the Xtra-Base group (0.88) and the lowest in the Aelite group (0.82). In the 4-mm groups, the highest HR was observed in the SDR and Xtra-Base groups (0.80) and the lowest in the Grandio and Aelite groups (0.73) [Table 3].

**DISCUSSION**

This study used three different bulk-fill flowable composites and two different methacrylate-based flowable composites with two different adhesive systems. The dentin bond strength of composite resin was evaluated using the \( \mu \)TBS test method and the HR of composite was measured using the Vickers microhardness measurement method.

The \( \mu \)TBS test was introduced by Sano et al.[9] in 1994. This method offers a number of advantages, such as providing a large amount of microspecimens, the ability to prepare microspecimens from different regions on a tooth, and the possibility of working in irregular surfaces.[29] The \( \mu \)TBS test was used in this study due to these advantages.

The Aelite and SDR etch-and-rinse groups exhibited statistically significantly higher bond strength (12.77 and 20.36 MPa, respectively) than the self-etch group (16.04 and 17.09 MPa, respectively) (\( P < 0.05 \)).
No statistically significant differences were determined between the X-Tra-Base, Grandio, and Filtek etch-and-rinse and self-etch groups ($P > 0.05$). Among the self-etch groups, there was a significant difference between Aelite and Filtek and the SDR and Xtra-Base groups. Among the etch-and-rinse groups, there was a significant difference between Aelite and the SDR groups. Our first hypothesis that there would be no differences in dentin bond strength between bulk-fill and methacrylate-based flowable composites was thus partially rejected.

The mean bond strengths of the etch-and-rinse groups were higher than those of the self-etch groups. A number of studies have compared the bond strengths of two-step etch-and-rinse and self-etch adhesives. Armstrong et al. reported that two-step etch-and-rinse adhesives have a higher dentin bond strength than two-step self-etch adhesives. Similarly, Villela-Rosa et al. showed that two-step etch-and-rinse adhesives have higher dentin bond strengths than self-etch adhesives. Studies have attributed these findings to the insufficient etching effect of self-etch adhesive and the poor penetration of its monomers to the dentin tubules. The lower mean bond strength in self-etch groups in our study may be ascribed to a similar mechanism. Increasing bond strength concerns not only adhesive structure, but also the chemical composition of composite resin. Differences in the chemical structures of composite resins may lead to different bond strengths during the application of adhesive resin. No statistically significant difference between the Xtra-Base, Grandio, and Filtek etch-and-rinse and self-etch groups in our study could be explained by the chemical structure of these composites.

The highest bond strength was observed in the SDR etch-and-rinse group. This may be related not only to SDR bulk-fill flowable composite’s monomer chemistry, but also to its surface energy and wettability characteristics which are higher than those of the other composites used. Van Ende et al. reported that bulk-fill flowable composites exhibited better internal adhesion than methacrylate-based flowable composites with higher wettability in cavities with high C-factor. Adaptation of the composite resin is associated with increased surface energy and the high surface energy effects of enhancing the bonding strength. Similarly, Moorthy et al. reported that bulk-fill composites have higher cavity adaptation than methacrylate-based composites.

Aelite flow exhibited the lowest bond strength in both the etch-and-rinse and self-etch groups. Park et al. investigated the bond strength of flowable composites and determined that Aelite flow specimens had lower bond strength than Grandio flow specimens. Studies have attributed this to the low filler content of the Aelite flow composite. Similarly, the lowest bond strength being determined in the Aelite flow group in our study may be due to its low filler content.

In this study, the surface microhardness of composite resins was evaluated using the Vickers microhardness measurement method. The tip of the Vickers microhardness tester is shorter than those of other hardness measurement devices. This allows the device to perform more accurate measurements and mean that it is less affected by the surface properties of the material. We therefore used a Vickers microhardness tester to measure microhardness.

Since chemical composition and filler content influence physical properties, differences may be observed between the microhardness values of composite resins. According to Braem et al., composite resins with a high filler content have higher surface microhardness. Similarly, Chung et al. showed the relationship between mechanical properties of composite resin and the amount of filler that surface microhardness decreases as the filler content in composite resins decreases. In our study, microhardness measurement results were similarly proportional to the amounts of composite filler. Surface microhardness values, from high to low, were observed in methacrylate-based Grandio Flow (81% by volume), bulk-fill composite Xtra-Base (58% by volume), SDR (44% by volume), Filtek (42.5% by volume), and methacrylate-based composite Aelite Flow (42% by volume). Salerno et al. reported that bulk-fill flowable composites have lower surface microhardness compared to methacrylate-based flowable composites. They attributed this to the lower filler amount in bulk-fill flowable composites.

Due to its low cost and ease of application, microhardness testing is the most commonly used method for measuring the degree of polymerization of composite resins. In this method, the HR is calculated by measuring the bottom and top surface microhardness. According to Bouschlicher et al., a HR of 0.80 or above is necessary for the sufficient polymerization of composite resin. In this study, HRs in all groups were above this in 2-mm specimens, while in 4-mm specimens, this level was achieved only in the Xtra-Base and SDR groups. Our second hypothesis, that bulk-fill composites would have a sufficient degree of polymerization at a 4-mm thickness, was thus also partially rejected.

El-Damanhoury and Platt reported in their study of the degree of polymerization of methacrylate-based and bulk-fill flowable composites that a HR of 0.80 was determined only in SDR and Xtra-Base groups in 4-mm thickness specimens. Methacrylate-based flowable composites used in that study did not exhibit a sufficient degree of polymerization. Similarly, Jang et al. also reported that methacrylate-based composites did not demonstrate a sufficient degree of polymerization at a 4-mm thickness.
The improved translucent structure of bulk-fill flowable composites and the photoactive groups inserted into methacrylate resin allows the composites to achieve better control of polymerization kinetics.[5] Manufacturers modified the chemistry of a monomer known as Bowen monomers (Bis-GMA: 2,2-bis-[4-(2-hydroxy-3-methacryloxypropoxy) phenyl] propane) to obtain a new low-viscosity monomer. In addition to this monomer, hydroxyl-free Bis-GMA, aliphatic urethane dimethacrylate, partial aromatic urethane dimethacrylate, or high-branched methacrylate have also been added to the bulk-fill flowable composite resin matrix.[22] This change in the organic matrix and monomer structure improves the translucent structure of bulk-fill flowable composite, permits light to spread further throughout the mass of the composite, and allows it to polymerize up to 4 mm in thickness.[23] Bulk-fill flowable composites thus exhibit a higher degree of polymerization to that of methacrylate-based flowable composites. The higher degree of polymerization of bulk-fill flowable composites in this study may also be attributed to this mechanism.

CONCLUSIONS

Within its limitations, this in vitro study shows that bulk-fill flowable composites exhibit a higher degree of polymerization than methacrylate-based flowable composites at a 4-mm thickness. During posterior restorations, bulk-fill flowable composites may be applied to cavities in layers >2 mm using a bulk technique.

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Conflicts of interest

There are no conflicts of interest.

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