Enhancement of Adhesion between Resin Coating Materials and Resin Cements

Tomoaki UDO1, Toru NIKAIDO1, Masaomi IKEDA1, Dinesh S WEERASINGHE2, Naoko HARADA2, Richard M. FOXTON3 and Junji TAGAMI2

1Cariology and Operative Dentistry, Department of Restorative Sciences, Graduate School, Tokyo Medical and Dental University, 5-45, Yushima 1-chome, Bunkyo-ku, Tokyo 113-8549, Japan
2Division of Conservative Dentistry, Kings' College London Dental Institute at Guy's, Kings' and St. Thomas' Hospitals, Kings' College, London, SE1-9RT, UK
3Center of Excellence Program for Frontier Research on Molecular Destruction and Reconstruction of Tooth and Bone, Tokyo Medical and Dental University, 5-45, Yushima 1-chome, Bunkyo-ku, Tokyo 113-8549, Japan

Corresponding author, Tomoaki UDO; E-mail: uddy0901@hotmail.com

Received December 22, 2006/Accepted March 2, 2007

Resin coating technique is a unique method that improves the dentin bond strength of resin cements in indirect restorations. However, the weak link of a specimen bonded using the resin coating technique was reported to be the bonded interface between the resin coating material and resin cement. The purpose of this study, therefore, was to enhance the bonding performance between a resin coating material and a resin cement. Two light-cured flowable composites, Protect Liner F and Clearfil Flow FX, were used as coating materials, and two dual-cure composite materials, Panavia F 2.0 and Clearfil DC Core Automix, were used as resin cements. The ultimate tensile strength of each material and the microtensile bond strengths of the bonded specimens of resin coating material and resin cement were measured using a crosshead speed of 1.0 mm/min. Three-way ANOVA (p<0.05) revealed that the highest microtensile bond strength was obtained using a combination of Clearfil Flow FX and Clearfil DC Core Automix, and when the surface of the coating material was treated with ED Primer II. It was strongly suggested that materials with a higher ultimate tensile strength, when used in both resin coating and cementation, could enhance the bond strength between the two.

Keywords: Resin coating, Resin cement, Pretreatment

INTRODUCTION

Tooth-colored, indirect restorations have become widely accepted in clinical practice, largely due to the vast improvements in adhesive materials and techniques. On this note, resin cements are used as an adhesive material for indirect restorations. However, currently available resin cements do not always bond to dentin as strongly as adhesive systems for direct resin composite restorations. It is noteworthy that a relatively weak bond to tooth structure leads to a host of problems: gap formation at the margin of restorations, postoperative sensitivity, premature failure, and secondary caries formation. Therefore, it is plain that there is an urgent, imperative need to improve the adhesive property of resin cements to dentin.

In a bid to improve dentin bond strength, a resin coating technique has been advocated since the early 1990s. By means of a combined use of a dentin adhesive system and a low-viscosity flowable resin composite, this technique produces a hybrid layer and a tight sealing film on the dentin surface. In this manner, the prepared dentin is well covered and protected immediately after preparation. This also means that pulpal irritation — which is caused by mechanical and thermal stimuli and bacterial infiltration during impression taking, temporization, and final cementation — is kept to a minimum. Other accompanying advantages of this technique include increased bond strength of resin cements, better sealing and adaptation to dentin, as well as inhibitory effect on coronal leakage in endodontically treated teeth.

It has been shown that with an optimal combination of a dentin bonding system and a low-viscosity flowable composite, a resin coating indeed enhanced the dentin bond strength of resin cements in indirect restorations. Nonetheless, the bond strength of indirect resin composite restorations is still lower than that of direct composite restorations. For debonded specimens of resin coating technique, SEM examination of the failure modes revealed that failure occurred at the interface between the resin cement and resin-coated surface. In other words, when a specimen is bonded using the resin coating technique, adhesion is weakest at the interface between the resin cement and resin coating material. Therefore, if adhesion of resin cement to the resin-coated surface were improved, the bond strength of the resin-coated specimen would be enhanced. On this note, the purpose of this study was to investigate how to enhance the bond strength of resin cement to the resin coating material.

The hypothesis of this study was that the use of coating materials and resin cements with higher ultimate tensile strength would increase the microtensile bond strength of resin cement to the
Adhesion to a resin-coated surface

resin coating material.

MATERIALS AND METHODS

Ultimate tensile strength measurement

Table 1 lists the materials used in this study: two flowable resin composites (Protect Liner F and Clearfil Flow FX, Kuraray Medical, Tokyo, Japan), a resin cement (Panavia F 2.0, Kuraray Medical), and a dual-cure composite core material (Clearfil DC Core Automix, Kuraray Medical). To measure the ultimate microtensile strength of each material, they were inserted into stainless steel molds (10 mm diameter, 4 mm height), pressed with a plastic matrix strip to flatten the surface, and light-cured for 40 seconds from the top surface using a light curing unit (Optilux 500, Demetron, Danbury, IL, USA). They were then sectioned into rectangular sticks (1×1×4 mm) using a low-speed diamond disk (Isomet 1000, Buehler, Lake Bluff, IL, USA). Fourteen rectangular sticks were made from each specimen. The specimens were then stressed in tension at a crosshead speed of 1 mm/min using a universal testing device (EZ-test, Shimadzu, Kyoto, Japan) to measure the ultimate tensile strength (UTS).

Independent t-test was used to compare the UTSs between Protect Liner F and Clearfil Flow FX, and between Panavia F 2.0 and Clearfil DC Core Automix, respectively (p=0.05).

Microtensile bond strength measurement

Specimen preparation is illustrated in Fig. 1. A resin composite disc (Clearfil AP-X, Shade A2, Kuraray Medical) was used as a substitute material for dentin in this study. The resin composite was inserted into a stainless steel mold (10 mm diameter, 2 mm height), pressed with a plastic strip to make a flat surface, and then light-cured for 40 seconds from the top surface using a light curing unit. Following which, one of two flowable resin composites, Protect Liner F or Clearfil Flow FX, was inserted into the stainless steel mold (300 μm thick) and placed on top of the cured composite, then pressed with a plastic strip and light-cured for 20 seconds. Surface of the cured flowable resin composite was wiped with a cotton pellet soaked in alcohol for 10 seconds to remove the unpolymerized layer on the surface. The specimens were then stored in 37°C water for one week.

To clean up the surfaces of the specimens, 37% phosphoric acid gel (K-etchant, Kuraray Medical) was applied for 10 seconds, rinsed thoroughly with a water spray for 10 seconds, and gently air-dried for

Table 1 Materials used in this study

<table>
<thead>
<tr>
<th>Material, (Shade)</th>
<th>Batch number</th>
<th>Composition</th>
<th>Filler content (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Protect Liner F</td>
<td>00057A</td>
<td>Bis-GMA, TEGMA, microfillers, photoinitiator</td>
<td>31</td>
</tr>
<tr>
<td>Frow FX</td>
<td>0001AE</td>
<td>Bis-GMA, TEGDMA, UDMA, microfillers, photoinitiator</td>
<td>65</td>
</tr>
<tr>
<td>Panavia F 2.0</td>
<td>0003B</td>
<td>Oxyguard II: Polyethylene glycol/glycerin gel</td>
<td>78</td>
</tr>
<tr>
<td>Clearfil DC Core Automix</td>
<td>0001B</td>
<td>Bis-GMA, TEGMA, hydrophobic aromatic dimethacrylate, silanated glass, silanated colloidal silica, di-Camphorquinone, benzoylperoxide</td>
<td>74</td>
</tr>
<tr>
<td>Clearfil SE Bond</td>
<td>00091B</td>
<td>A: MDP, HEMA, 5-NMSA, chemical initiator</td>
<td>65</td>
</tr>
<tr>
<td>ED primer II</td>
<td>0009B</td>
<td>B: 5-NMSA, water</td>
<td>65</td>
</tr>
<tr>
<td>Porcelain Bond Activator</td>
<td>00142A</td>
<td>Bis-PEDMA, MPS</td>
<td>65</td>
</tr>
<tr>
<td>ES(DA3)</td>
<td>D13-CB-Ti</td>
<td>Urethane monomer, Alumina ultrafine filler</td>
<td>92</td>
</tr>
</tbody>
</table>

Manufacturer: Kuraray Medical, Tokyo, Japan

Bis-GMA: bisphenol-A-diglycidylmethacrylate; TEGMA: triethylene glycol dimethacrylate; UDMA: urethane dimethacrylate; BFEDMA: bisphenol-A-polyethoxy dimethacrylate; MDP: 10-methacryloyloxydecyl dihydrogenphosphate; DMA: aliphatic dimethacrylate; Ba: barium; B: boron; Si: silica; HEMA: hydroxyethyl methacrylate; 5-NMSA: N-methacryloyl-5-aminosalicylic acid; Bis-PEDMA: bisphenol-A-polyethoxy dimethacrylate; MPS: 5-methacryloyloxy propyl trimethoxysilane
10 seconds. Following which, they received one of two surface pretreatments: no treatment or application of ED Primer II (Kuraray Medical) for 30 seconds and gently air-blown for five seconds.

To simulate the bonding of an indirect composite restoration, indirect composite slabs (2 mm thick, 10 mm diameter) were fabricated from Estenia (DA2, Kuraray Medical). The fabrication process entailed light curing for three minutes using a light curing unit (Alpha Light II, J. Morita, Tokyo, Japan), followed by heat curing at 110°C for 15 minutes in an oven (KL-100, Kuraray Medical) according to manufacturer's instructions.

The indirect composite disk was then bonded to the resin-coated surface using one of the two materials: dual-cure resin cement (Panavia F 2.0, Kuraray Medical) or dual-cure composite core (Clearfil DC Core Automix, Kuraray Medical). Surfaces of the Estenia slabs to be bonded were etched with 37% phosphoric acid gel (K-etchant gel) for 30 seconds, rinsed, and dried. Then, the surface was silanized with a mixture of SE Primer of Clearfil SE Bond (Kuraray Medical) and Clearfil Porcelain Bond Activator (Kuraray Medical) and gently air-dried. Half of the Estenia slabs were cemented with Panavia F 2.0 and light-cured for 40 seconds. The other half of the specimens were cemented with Clearfil DC Core Automix and light-cured for 40 seconds.

Bonded specimens were stored in water at 37°C for one day. They were then sectioned into rectangular sticks (1×1×5 mm) using a low-speed diamond disk. Six rectangular sticks were made from each specimen. The specimens were then fixed to Ciucchi's jig with cyanoacrylate glue (Model Repair II Blue, Dentsply-Sankin, Ohtawara, Japan) and stressed in tension at a crosshead speed of 1 mm/min using a universal testing device (EZ-test).

Mean microtensile bond strengths (μTBS) were statistically analyzed by three-way ANOVA. The factors analyzed were resin coating material, pretreatment of coating material, and type of resin cement. After eliminating the significant interactions of three-way ANOVA and two-way ANOVA, Dunnett's T3 test was used to compare the effects of the types of resin coating material and resin cement.
on µTBS (p=0.05).

Fracture mode examination
To examine the fracture modes of the debonded specimens after microtensile bond testing, the fractured specimens were gold sputter-coated and observed under a field emission scanning electron microscope (FE-SEM; S4500, Hitachi, Tokyo, Japan). The fracture modes were classified into two types as shown in Fig. 2: Type 1—Complete or partial adhesive failure at the interface, including cohesive failure in the resin cement or resin coating material; Type 2—Complete cohesive failure in the resin cement. Results of the fracture modes were analyzed using Fisher’s exact test at 95% level of confidence.

RESULTS
Ultimate tensile strength
Table 2 shows the ultimate tensile test results of the flowable composites and the composite materials for cementation. Independent t-test revealed that the UTS of Clearfil Flow FX was significantly higher than that of Protect Liner F, while the UTS of Clearfil DC Core Automix was significantly higher than that of Panavia F 2.0 (p<0.001).

Microtensile bond strength
Table 3 lists the mean microtensile bond strengths of the resin cements to the resin coating materials. Three-way ANOVA indicated a significant interaction between pretreatment and resin cement (p=0.001), while no interactions were found between resin coating material and pretreatment (p=0.756) and between resin coating material and resin cement (p=0.910). Following this, two-way ANOVA was carried out separately with two factors, Protect Liner F and Clearfil Flow FX.

Two-way ANOVA indicated significant interactions between pretreatment and resin cement in Protect Liner F (p=0.021) and Clearfil Flow FX.

Table 2 Ultimate microtensile strengths of coating materials and resin cements (MPa)

<table>
<thead>
<tr>
<th>Material</th>
<th>Tensile Strength</th>
</tr>
</thead>
<tbody>
<tr>
<td>Protect Liner F</td>
<td>93.3 ± 17.8a</td>
</tr>
<tr>
<td>Flow FX</td>
<td>170.1 ± 22.7b</td>
</tr>
<tr>
<td>Panavia F2.0</td>
<td>57.15 ± 15.11c</td>
</tr>
<tr>
<td>DC Core Automix</td>
<td>159.19 ± 19.8d</td>
</tr>
</tbody>
</table>

Number of specimens: 10, Mean ± SD
Same superscript letter indicates statistically similar groups (p<0.05)

Table 3 Effects of coating material and resin cement on microtensile bond strength (MPa)

<table>
<thead>
<tr>
<th>Coating material</th>
<th>Pretreatment</th>
<th>Resin cement</th>
</tr>
</thead>
<tbody>
<tr>
<td>Protect Liner F</td>
<td>None</td>
<td>Panavia F2.0</td>
</tr>
<tr>
<td></td>
<td>ED Primer II</td>
<td>DC Core Automix</td>
</tr>
<tr>
<td>Clearfil Flow FX</td>
<td>None</td>
<td></td>
</tr>
<tr>
<td></td>
<td>ED Primer II</td>
<td></td>
</tr>
</tbody>
</table>

Number of specimens: 30, Mean ± SD
Same superscript letter indicates statistically similar groups (p<0.05)

Table 4 Effects of resin cement on fracture mode after microtensile bond testing

<table>
<thead>
<tr>
<th>Coating material</th>
<th>Pretreatment</th>
<th>Resin cement</th>
<th>Type I</th>
<th>Type II</th>
</tr>
</thead>
<tbody>
<tr>
<td>Protect Liner F</td>
<td>None</td>
<td>Panavia F2.0</td>
<td>27</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>ED Primer II</td>
<td>DC Core Automix</td>
<td>21</td>
<td>9</td>
</tr>
<tr>
<td>Clearfil Flow FX</td>
<td>None</td>
<td>Panavia F2.0</td>
<td>27</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>ED Primer II</td>
<td>DC Core Automix</td>
<td>18</td>
<td>12</td>
</tr>
</tbody>
</table>

Adhesion to a resin-coated surface

Fig. 3 FE-SEM images of the fractured surfaces: (a) Type 1; (b) Type 2.
(p=0.015), respectively. Dunnett’s T3 test revealed that the microtensile bond strengths of ED Primer II were significantly higher than those without pretreatment for each cement (p<0.05). The microtensile bond strengths of Clearfil DC Core Automix were significantly higher than those of Panavia F 2.0, when the coating material was pretreated with ED Primer II (p<0.05). However, there were no significant differences in bond strength between Panavia F 2.0 and Clearfil DC Core Automix, when the coating material was not pretreated (p>0.05).

Highest bond strength was obtained with the combination of Clearfil Flow FX and Clearfil DC Core Automix, when the surface of the coating material was treated with ED Primer II. However, this value was not statistically different from that with the combination of Protect Liner F and Clearfil DC Core Automix (p>0.05).

Fracture modes

Table 4 lists the fracture modes for each group. SEM pictures of the typical fracture modes are shown in Fig. 3. Type 1 failure was more obvious than Type 2 in all the groups. Fisher’s exact test revealed that there were no significant differences in fracture mode between no-pretreatment and pretreatment with ED Primer II, except for the combination of Clearfil Flow FX and Panavia F 2.0 (p>0.05), whereby pretreatment of the resin coating material with ED Primer II increased the incidence rate of Type 2 failure.

DISCUSSION

In clinical practice, the resin coating technique is highly recommended for indirect inlay/onlay restorations. This technique entails the combined use of a dentin bonding system and a flowable resin composite on the prepared dentin surface. A single coating of a dentin bonding system can protect the prepared dentin and minimize pulp irritation. However, the additional use of a flowable resin composite followed by the dentin bonding system demonstrated significantly higher bond strengths of the resin cement to dentin compared with a single coating of the dentin bonding system. This is chiefly because the flowable resin composite plays multiple beneficial roles: enhance subsequent polymerization of the oxygen inhibition layer in the uncured adhesive resin, protect the cured adhesive from being torn at the time of temporary cement removal, as well as function as a stress breaker.

Nonetheless, to achieve the desired bond strength between a resin cement and the resin-coated dentin, one key factor is the careful and appropriate selection of the adhesive system. It was reported that the combination of Clearfil SE Bond and Protect Liner F provided the highest bond strength of Panavia F to dentin. However, the resin cement bond strength was still lower than that of direct composites. On this account, fracture modes of the debonded specimens after bond strength testing suggested that the weak link of bonded specimens — by resin coating technique — was the bonded interface between the resin coating material and the resin cement. In light of this finding, we focused on improving the interface between them.

A resin composite, Clearfil AP-X, was used as a substitute material for dentin substrates in this study. Clinically, the resin coating technique is applied to the prepared dentin surface. However, dentin is not a homogeneous substrate, which may influence the microtensile bond strength, and hence the fracture mode, of debonded specimens. To preclude the multifaceted influences of dentin which may complicate test results, a resin composite was used instead in this study as it is more uniform than dentin. In addition, tight bonding between Clearfil AP-X and the applied coating material can be obtained because of their similar compositions.

Two flowable composites, Protect Liner F and Clearfil Flow FX, were compared as coating materials. Protect Liner F was developed as a lining material for direct resin composites and has been introduced as a coating material since the early 1990s. Recently, various flowable resin composites with different filler loads and viscosities have been marketed. Clearfil Flow FX is a flowable resin composite with a multi-purpose use. The indications for a flowable resin composite are direct filling of small cavities without a strong stress-bearing area. Protect Liner F and Clearfil Flow FX are both Bis-GMA-based, but the filler load of Clearfil Flow FX is comparatively higher than that of Protect Liner F. The present study demonstrated that the ultimate tensile strength of Clearfil Flow FX was significantly higher than that of Protect Liner F.

After light-curing the resin coating material, the top surface of the coating material was wiped with a cotton pellet soaked in alcohol to remove the unpolymerized layer. The unpolymerized layer remaining on the resin coating material may be beneficial in helping to bond resin cement to the coating material. However, the unpolymerized layer affects the setting reaction of impression materials — especially silicone impression materials, resulting in poor dimensional accuracy of the cast model. In light of this concern, the unpolymerized layer should be wiped off — which may then result in relatively lower bond strengths of resin cement to the resin coating material as compared to dentin.

Before cementation, the manufacturer recommended treating the coating surface with ED Primer...
II, which contains MDP, HEMA, 5-NMSA, chemical initiation, and water. ED Primer II improves surface wettability and promotes interfacial polymerization. Results of the present study clearly demonstrated that application of ED Primer II indeed significantly increased the bond strength of the resin cement to the coating material in all the groups.

Panavia F 2.0 and Clearfil DC Core Automix were used for the cementation of indirect composites in this study. Panavia F 2.0 is a dual-cure resin cement, which contains MDP as a component. Clearfil DC Core Automix was originally used as a composite core material. However, Clearfil DC Core is also a dual-cure material. On this account, some researchers have used this material for the cementation of indirect restorations. Panavia F 2.0 contains 78 wt% filler loading, while Clearfil DC Core Automix has 74 wt% fillers. However, Clearfil DC Core Automix exhibited a higher ultimate tensile strength than Panavia F 2.0, which might be due to a different composition of the resin matrix. It was reported that resin cements with higher ultimate tensile strengths produced higher bond strengths to human dentin. However, it should be noted that there was no correlation between the ultimate tensile bond strength and filler loading of resin cements.

Based on the failure mode results, it could be seen that Type 1 failure was chiefly observed in all the groups, except for the combination of Clearfil Flow FX and Panavia F 2.0. Therefore, the weak link was at the interface between the resin coating material and resin cement. On the other hand, pretreatment of the resin coating material with ED Primer II remarkably increased the incidence rate of Type 2 failure in the combination of Clearfil Flow FX and Panavia F 2.0. While the reason for this behavior remained unclear, it was speculated to be related to the polymerization behavior of Panavia F 2.0 in the presence of ED Primer II.

Results of the current study showed that when materials with a higher ultimate tensile strength were used in resin coating and cementation, enhanced bond strength between the resin coating material and resin cement was yielded. In particular, highest bond strength was obtained with the combined use of Clearfil Flow FX and Clearfil DC Automix, and when the surface of the coating material was treated with ED Primer II. Based on this result, the hypothesis of this study was accepted.

Dentin bonding performance of direct composites is still more reliable than that of a resin cement with indirect composites. Caries treatment with minimal intervention has been achieved in direct composite restorations, but not so in indirect restorations. Cavity preparations for inlay/onlay restorations are still based on GV Black’s principles, whereby sound dentin is cut to obtain retention and resistance forms. However, resistance and retention forms are not required if good bonding to dentin can be achieved. On this note, we proposed the use of the resin coating technique for indirect composite restorations.

Our previous study reported that the resin coating technique increased the bonding performance of resin cements to dentin remarkably. This finding strongly suggested the possibility of employing minimally invasive cavity preparations for indirect restorations. Moreover, the present study demonstrated that the bond strength of a resin cement to a resin coating material could be enhanced, whereby a resin composite was used as a substitute material for dentin substrate. Therefore, results of the present study paved the way for more combinations of resin cement and resin coating material to be explored, and their effects on dentin bond strength evaluated to the end of achieving improved bond strength for clinical use.

CONCLUSIONS

Within the limitations of this study, materials with higher ultimate tensile strength used in resin coating and cementation enhanced the bond strength between the resin coating material and resin cement. Highest bond strength was obtained with the combination of Clearfil Flow FX and Clearfil DC Core Automix, and when the surface of the coating material was treated with ED Primer II.

ACKNOWLEDGEMENTS

This work was supported by a grant from the Center of Excellence Program for Frontier Research on Molecular Destruction and Reconstruction of Tooth and Bone, Tokyo Medical and Dental University.

REFERENCES

6) Islam MR, Takada T, Weerasinghe DS, Uzzaman MA,


