Over-etching effects on micro-tensile bond strength and failure patterns for two dentin bonding systems

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Received 21 April 2001; revised 22 September 2001; accepted 31 October 2001

Abstract

Objectives. The purpose of this study was to determine (1) the weakest zone of resin–dentin bonds and (2) the relation between bond strength and failure mode to clarify the effect of demineralized dentin.

Methods. Human premolars were sectioned to expose the dentin surfaces, and the dentin surfaces were conditioned with phosphoric acid for 15, 60, 120, or 180 s. Resin–dentin bonded specimens were produced using two adhesives: One-Step (Bisco) and OptiBond Solo (Kerr). Each sample was sectioned to produce a beam (adhesive area: 0.9 mm²). Microtensile bond tests were then conducted, and the mean bond strengths (n = 12 for each group) were statistically compared using two-way ANOVA and Duncan’s multiple-range test (p < 0.05). The fractured surfaces of all specimens were examined using SEM, and the areas of failure were measured using an image analyzer.

Results. For One-Step, the bond strength decreased with increase in acid-conditioning time (15 s: 50.7 ± 9.7, 60 s: 40.8 ± 11.0, 120 s: 23.6 ± 4.9 and 180 s: 12.1 ± 4.6 MPa) (p < 0.05). For OptiBond Solo, the bond strength in the case of 15 s acid-conditioning time (42.6 ± 7.9 MPa) was significantly greater than that for the other times (60 s: 31.9 ± 10.3, 120 s: 31.8 ± 14.4 and 180 s: 31.8 ± 7.4 MPa) (p < 0.05). Fractography showed that the area percentage of the hybrid layer increased with increase in etching time for both systems.

Conclusions. The integrity of the hybrid layer, especially the top part, has an effect on bond strength.

Keywords: Fractography; Micro-tensile bond test; Hybrid layer; Demineralized dentin

1. Introduction

The recent development of functional monomers in bonding resin systems has improved the integrity of resin–dentin bond structures. However, the presence of demineralized dentin within the bonded interface has been reported under SEM observation. This demineralized dentin zone within the bonded structure is created by incomplete resin diffusion of the collagen meshwork after acid-etching [1–6]. It has been suggested that the degree of resin infiltration of the exposed collagen fibrils within the demineralized dentin has a profound influence on bond integrity [7–9]. Furthermore, depletion of the collagen fibrils within the demineralized dentin zone was observed at the fractured surface using human substrates in a long-term in vivo study [6]. These previous reports suggested that the presence of this demineralized dentin zone might reduce the bond strength. Thus, the demineralized dentin zone within the bond structure has lately become a subject of special interest in dentistry.

Using SEM, TEM and fractographic analysis, several reports showed controversial results indicating that either the top of the hybrid layer [10–12] or the demineralized dentin zone [9,13] could be the weakest link within the bonded interface. However, little is known about the relationship between the failure mode (the area percentage of the failure) and bond strength.

Based on the assumption that excessive acid-conditioning for dentin pretreatment would form a deep demineralized dentin zone within the bonded structures, we used specimens that had been exposed to prolonged acid-conditioning to evaluate the relation between the demineralized dentin and bond strength. Therefore, the objectives of
this study were: (1) to evaluate the bond strengths of two single bottle adhesives that had been acid-conditioned for different periods to dentin, and (2) to relate these bond strengths with the distribution of failure modes as observed by SEM. The null hypothesis tested was that there is no difference in resin–dentin bond strength and location of bond failures in dentin regardless of etching time.

2. Materials and methods

2.1. Test design

This test was designed to determine the relationship between bond strength and failure patterns of demineralized dentin by using an over-etching technique to artificially produce the demineralized dentin in the bond structures.

2.2. Specimen preparation

Human premolars, extracted for orthodontic reasons with the informed consent of the patients, were used in this study. They were stored in normal saline solution at 4 °C and bonded within 2 weeks of extraction. Flat dentin surfaces were created by removing the enamel and superficial dentin by sectioning the crown (mid-coronal portion) at right angles to the long axis of the teeth using a water-cooled low-speed diamond saw (Isomet; Buehler Ltd, Lake Bluff, IL). The surface was then ground with 600-grit silicon carbide paper under water to create a uniform smear layer [14].

2.3. Bonding procedure

Two commercially available adhesive resin systems, One-Step (OS: Bisco Inc., Schaumburg, IL) and OptiBond Solo (OP: Kerr Corp., Orange, CA) were investigated in this study (Table 1). The prepared dentin surfaces were randomly assigned into eight groups and acid-conditioned for 15, 60, 120, or 180 s with either OS or OP. The acid-conditioning time recommended by the manufacturer was 15 s for each system.

Excess water was blot-dried from the dentin surface with a cotton pellet, leaving the surface visibly moist. At least two coats of the bonding resin were then consecutively applied and dried with oil-free compressed air using an air syringe. Subsequently, the surface was light-cured for 10 s with a light-curing unit (Curing Light XL 3000; 3M, St Paul, MN). The bonded surfaces were examined with a magnifying lens to ensure that bonded surfaces were shimmer prior to light-activation. When a matte surface was observed, additional coats of adhesive were applied. After applying the bonding resin, four 1 mm thick increments of the resin composite were built up and light-cured for 60 s for each increment.

2.4. Microtensile bond test

The bonded specimens were stored in sterilized water at 37 °C for 24 h. Each specimen was sectioned perpendicular to the adhesive interface with an Isomet diamond saw under wet conditions to remove a slab from the center of the bonded tooth (Fig. 1). This resin–dentin bonded slab was further sectioned to produce beams, with adhesive areas being an average of 0.9 mm² (Fig. 1) [15]. Four beams per tooth were obtained. In each of the eight groups, 12 beams
obtained from three teeth were tested. Each beam was then attached to a testing apparatus with a cyanoacrylate adhesive (Model Repair II blue, Sankin Industry Co., Ltd, Tokyo, Japan) and a tensile load was applied with a material tester (EZ Test, Shimadzu Co., Kyoto, Japan) at a crosshead speed of 1.0 mm/min. Twelve beams were tested from each of the eight groups. The mean bond strengths were statistically analyzed by two-way ANOVA and Duncan’s multiple-range test. The two variables compared were tensile bond strength and acid-conditioning time. Statistical significance was set in advance at the 0.05 probability level.

2.5. Fractographic analysis using SEM

After the microtensile test, fractured surfaces from the dentin sides of all specimens were sputter-coated with gold and observed with a field-emission scanning electron microscope (S-4000, Hitachi Ltd, Tokyo, Japan). The area fractions of each failure mode were expressed as percentages of the total fractured surfaces of all specimens. They were calculated from the SEM photographs using an image analyzer (Digitizer, KD4030B; Graphtec, Tokyo, Japan) to evaluate the failure pattern for each adhesive system. Failure modes of resin–dentin bonds were categorized into three types: failure in bonding resin or resin composite (I), in the hybrid layer (II), and in the demineralized dentin zone (III) (Fig. 2).

3. Results

Fig. 3 shows the tensile bond strengths for the eight groups (n = 12 for each group). For OS, the tensile bond strength decreased with increasing acid-conditioning time (15 s: 50.7 ± 9.7, 60 s: 40.8 ± 11.0, 120 s: 23.6 ± 4.9, 180 s: 12.1 ± 4.6 MPa) and the differences among these OS groups were statistically significant (p < 0.05). For OP, the bond strength for 15 s (42.6 ± 7.9 MPa) was significantly greater than for the other groups (60 s: 31.9 ± 10.3, 120 s: 31.8 ± 14.4, 180 s: 31.8 ± 7.4 MPa) (p < 0.05). However, there was no significant difference among the 60, 120, and 180 s groups (p > 0.05).

Fig. 4 shows the percentages of the failure modes observed for the two adhesives that were bonded using different etching times. In all groups, failure within the demineralized dentin represented only a minimal percentage of the total recorded failures. The area of the cohesive failure of the resin composite or the bonding resin decreased with increasing acid-conditioning time for OS. However, no
change in the area percentage was observed among the 60, 120, and 180 s groups for OP.

Fig. 5A is a SEM microphotograph showing the total fractured surface from the dentin side of a specimen in OP, in which the dentin was acid-conditioned for 60 s. The areas marked B and C in this figure are shown at higher magnification in (B) and (C). (B) Note the diagonal scratch lines creating the smear layer, indicating that failure occurred at the top of the hybrid layer. (C) Loss of peritubular dentin matrix and presence of collagen fibrils free of resin indicates this site as demineralized dentin at the base of the hybrid layer. EC, exposed collagen fibrils; T, fractured resin tag.

Fig. 6 presents the failure within the hybrid layer from the dentin (Fig. 6A) and resin sides (Fig. 6B) of a specimen in OP that was acid-conditioned for 180 s. Many short collagen fibrils were found protruding from the dentin side of the fractured beam (Fig. 6A). The irregular shadows are filler particles in the adhesive. The filler particles (white arrows) were scattered within the hybrid layer.

The total fractured surface from the dentin side of a representative specimen in OS that was acid-conditioned for 180 s is shown in Fig. 7A. The area marked B in the figure is shown at higher magnification in Fig. 7A. Failure at the top of the hybrid layer could be identified at the fracture surface.

4. Discussion

The classification of failure modes has significant implications in the present study. The relatively small surface area of specimens used in the microtensile bond test
enabled the percentages of failure modes to be documented more quantitatively for each specimen by using SEM observation [16]. The fractured surfaces shown in Figs. 5B and 7B both demonstrated scratches caused by polishing of the specimens. Their presence suggested that failure occurred along the boundary between the hybrid layer and the bonding resin layer. Conversely the fibrous nature of the intertubular dentin and the loss of the peritubular dentin in other specimens indicated that some fractures occurred in the demineralized dentin zone (Fig. 5C). This zone was created due to incomplete resin impregnation within the demineralized collage network.

The effect of acid-conditioning time on tensile bond strength (Fig. 3) is in good agreement with the percentage distribution of failure modes (Fig. 4). For One-Step, the failure zones (%) of the resin and the tensile bond strength were decreased with acid-conditioning time. For OptiBond solo, the failure zone (%) of resin and that of the hybrid layer were similar with 60, 120, and 180 s of acid-conditioning. This relation was similar to that between the tensile bond strength and the acid-conditioning time. The major area-fraction failure modes were indicative of the region with the highest stress concentration during testing, within the resin–dentin interface that was produced by acid-conditioning [13]. Several studies reported that prolonged acid-conditioning time resulted in fracture within the demineralized dentin zone when specimens were stressed to failure using the microtensile bond test [9,13]. This means that excessive acid-conditioning causes deeper demineralization of both intertubular and peritubular dentin, which in turn meant that they were not capable of being completely infiltrated by resin monomers. These studies suggested that the failure was likely to be initiated in this weakest zone, leading to decreased bond strength.

In contrast, microtensile bond strength studies using One-Step as the adhesive invariably showed that failure occurred on the top of the hybrid layer [10–12,17,18]. In the present study, this particular mode of failure for One-Step appeared to increase in proportion with the etching time (Fig. 4), from 36% when the manufacturer’s recommended time of 15 s was followed, to 82% when dentin was acid-conditioned for 180 s. In our two previous studies [12,17], the demineralized dentin was created by drying the acid-etched dentin before bonding. In this study, we created similar zones of incomplete resin infiltration by deliberately over-etching the dentin. The results of this work require rejection of the null hypothesis that there is no difference in the location of bond failures in dentin, regardless of etching time.

Van Meerbeek et al. [19] reported using the nano-indentation technique to evaluate the mechanical properties of resin–dentin bonds. In their research, the hardness and Young’s modulus of the hybrid layer were significantly different from those of the bonding resin. Under tensile tension, in the interface region of a bonded structure with two different components (such as the hybrid layer and the bonding resin layer) localized stress may be enhanced, leading to the propagation of a fracture at the boundary between the hybrid layer and bonding resin. This is one possible explanation for the results in the present study in which the major portion of the fractured surface was located at the top of the hybrid layer in both systems under excessive acid-conditioning.

A general trend that could be deduced from these studies is that the controversy appears to be adhesive system specific, and is probably related to the type of solvent employed in the adhesive system. When a water-free, acetone-based adhesive such as One-Step was applied either to briefly air-dried or over-etched dentin, failure occurred predominantly at the surface of the hybrid layer under tensile stress. Conversely, when water-containing, ethanol-based systems such as Single Bond (3M, Dental products, St Paul, MN) or OptiBond Solo were used, at least half of the failure modes were cohesive in nature within the zones of the demineralized dentin. TEM fractographic analysis of air-dried, bonded acid-etched dentin further showed that collagen fibrils were mechanically disrupted when failures...
occurred within the demineralized dentin. The torn edges of these fibrils exhibited unraveling of their subfibrillar architecture [12]. We speculated that if the fractured specimens in OptiBond Solo were examined using TEM, similar mechanical disruptions of the demineralized dentin would have occurred in over-etched dentin. This could be demonstrated, at least partially, in the SEM micrographs of the present study by the presence of short exposed collagen strands that were retained on the dentin sides of the fractured beams (Fig. 6A). On the other hand, when failure occurred at the surface of the hybrid layer, collagen fibrils within the hybrid layer were intact but demonstrated evidence of irreversible plastic deformation [12].

One possible way of explaining these observations is that an acetone solvent allows better affinity between the collagen fibrils and the resin because of its water-displacement potential. An ethanol/water solvent, by contrast, may produce weaker or no interaction between the surface of the collagen fibrils and the resin. Nakabayashi and Pasley [20] postulated three models (interface, interphase and denatured interphase model) to describe the potential interactions between demineralized collagen and polymerized resin within the hybrid layer. In the interface model, the resin envelops the fibril like a sheath without any micromechanical or chemical interaction. According to this model, collagen fibrils may be pulled out of the resin upon the application of tensile stress. The presence of collagen fibrils on the adhesive suggested that they plastically deformed during debonding. This deformation might have been due to the tensile stress. Such a scenario may occur when an ethanol/water solvent is employed. Thus, the morphological appearance shown in Fig. 6A was in accord with the interface model. In the interphase and denatured interphase model, resin interacts with collagen fibrils and forms more intimate contacts. The subfibrillar architecture of collagen fibrils contains spaces in which resin can potentially infiltrate [21,22]. This situation may be present when acetone is used as a resin solvent. The three hypothetical models have different implications when collagen fibrils are subjected to tensile stress. Whereas fibrils that are pulled out of resin in the interface model must bear the entire load during the final moment of breakage, both resin and collagen contribute to supporting the applied load in the interphase model or the denatured interphase model. The protective function of resin over collagen fibrils in the latter model is analogous to the mineral phase of the biological composite in sound dentin. This may partially explain the difference in failure modes observed in the adhesives examined.

The bond structure created with over-acid etching might have a different morphological appearance compared to that using the conditioning time recommended by the manufacturer. However, micromorphologically, it is possible that a similarly appearing structure was formed, regardless of acid-conditioning times. Extreme experimental conditions (such as the excessive acid-conditioning in this study) are useful in examining the adhesive structures formed during resin-bonding. Moreover, measuring the area percentage of the failure modes is an effective approach to evaluate resin–dentin bonds.

5. Conclusions

There are two main conclusions of this study.

1. The presence of a demineralized dentin zone has little adverse effect on the bond strength.
2. A relation was found between the bond strength and the area percentage of the failure mode of the resin and the hybrid layer.

Acknowledgements

This work was supported in part by a Grant-in-Aid for Scientific Research (No. 11470401) and by High Performance Biomedical Materials Research from the Ministry of Education, Science and Culture, Japan. The authors thank Mr Shuuichiro Hayashi of the Center for Electron Microscopy and Bio-Imaging Research of Iwate Medical University for a helpful discussion on FE-SEM.

References


