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Effect of Tubular Orientation on the Dentin Bond Strength of Acidic Self-etch Adhesives

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Clinical Relevance

The effectiveness of self-etch adhesive systems is dependent on both the physical properties of the adhesive and the integrity of the adhesive-dentin interface. Most importantly, the integrity of the adhesive-dentin interface is affected by the tubule orientation of the intaglio dentin structure. The initial bond strength and potential durability of the self-etch adhesive interfaces to dentin are significantly affected by both the adhesive pH and occlusally-oriented tubule direction. The clinical relevance to the dentist is that the results reported for bond strengths by both manufacturers and independent researchers should be interpreted and compared based on the orientation of the dentin used for the measurements, particularly for those adhesives that are very acidic (pH lower than 2).

ABSTRACT

OBJECTIVE: To determine the effect of varying the adhesive pH on the bond strength of an experimental self-etch adhesive system (BISCO, Inc) to dentin with the bonding surface perpendicular (occlusal direction) or parallel (axial direction) to dentin tubules.

METHODS: An experimental self-etch adhesive was modified by adding base to increase the pH from 1.1 to 2.7. Shear bond strength (SBS) was measured using an Ultradent jig method with Aelite All-Purpose Body light-cured composite (BISCO, Inc). Human dentin was prepared by exposing the axial and occlusal surface. The self-etch adhesive was applied according to the manufacturer's instructions and cured for 10 seconds@500mW using a VIP (BISCO, Inc) halogen curing light. Scanning electron microscope (SEM) examination was used to view both the occlusal- and axial-oriented dentin surfaces that were etched using pH (1.2~3.0) adjusted phosphoric acid solutions. All bond strength data analysis was performed using ANOVA, followed by a Student-Newman-Keuls multiple range test.

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DOI: 10.2341/10-233-LR

RESULTS: When the dentin-bonding surface was parallel to the tubule orientation (axial), the bond strength was independent of the pH of the self-etch adhesive (*p*>0.05). When the bonded surface was perpendicular to the tubule orientation (occlusal), the bond strength numbers were decreased as the pH decreased; the decrease became statistically significant when the pH was lower than 1.8. With a pH higher than 2.3, the bond strength had no difference (*p*>0.05) between the occlusal and axial positions. When the pH was lower than 1.8, SEM pictures confirmed that the smear layers and smear plugs were completely solubilized by the phosphoric acid solution. Higher pH values (2.0-2.8) showed smear layers partially solubilized and pH values of 3.0 fully retained the smear layer.

INTRODUCTION

Dentin is a complex biological structure with highly oriented microscopic channels, called dentinal tubules.[1-3] Dentin tubules continuously run to the pulp from the dentin-enamel junction (DEJ) and the CEJ in the root, with the wider portion oriented towards the pulp. Dentin tubules occupy approximately 1% of the total surface area at the DEJ and 22% near the pulp chamber.[4] Studies show that the orientation of the dentinal tubules has a great influence on the physical properties of dentin[5] and might also influence dentin bonding when using a total-etch technique.[4,6-14] The orientation of the dentin tubules has a great influence on the formation of the resin tags and hybrid layer.[7,13-14] When bonding to a dentin surface perpendicular to the tubule orientation (occlusal bonding), long and solid resin tags were observed; while when bonding to a surface parallel to the tubule orientation (axial bonding), no resin tags were formed. It is important to note that, on the occlusal surface, dentinal fluid within the occlusally-oriented tubules easily flows under pressure onto the cut/exposed dentin surface.[15-21] This fluid interferes with the quality of the adhesive-dentin interface and may decrease the bond strength between the resins and dentin (hydrophobic resins do not adhere to hydrophilic surfaces).[4]

Self-etch adhesives are specifically formulated to contain acidic monomers that serve to alter, rather than remove, the smear layer. These adhesives are hydrophilic and can create permeable membranes. Drs Tay and Pashley showed that aggressive self-etch adhesives completely (Prompt L-Pop, 3M ESPE, St Paul, MN, USA; pH 1.0) or partially (Non-rinse Conditioner/Prime & Bond NT, Dentsply, Milford, DE, USA; pH 1.2) solubilized smear layers and smear plugs, while, with mild self-etch adhesives (Clearfil SE Bond, Kuraray Medical Inc, Kurashiki, Okayama, Japan; pH 2.0), the smear layers and smear plugs were retained.[22] Removal or solubilizing of the smear layers/plugs with acidic adhesive/solutions (aggressive self-etch adhesive) opens up the dentin tubules, resulting in increased dentin surface wetness, which might decrease the dentin bond strength, especially for occlusal bonding (perpendicular to the tubules). Recent studies by Tay and others on the physical structure of cured self-etch adhesives have shown the influence of water migration through the dentin tubules, resulting in water tree formation and loss of bond strength.[17-18] The acidity of the adhesive layer seems to be related to the ease of diffusion of water through the adhesive, causing a loss of bond strength, particularly for self-cured dental materials. Further investigation of the influence of pH on adhesive and dentinal tubule orientation in regards to bond strength is needed in order to gain a better understanding of bonding mechanisms and durability.

The following study investigated the relationship between the pH (acidity) of a self-etching adhesive and dentin tubule orientation (perpendicular and parallel) and measured their effect on dentin bond strength.

METHODS AND MATERIALS

All materials were used according to the manufacturers' instructions. Experimental Self-Etch Adhesive (ExpSE, BISCO, Inc, Schaumburg, IL, USA) was formulated with varying pH and provided by BISCO, Inc. ExpSE includes an acidic phosphate monomer, resin monomers (bisphenol A diglycidyl methacrylate, 2-hydroxyl methacrylate), photo initiators (camphorquinone, amine) and solvent (water and ethanol). Potassium hydroxide (Sigma-Aldrich, St Louis, MO, USA, 99.99%) was used as received.

Human-dentin specimens were prepared by mounting teeth occlusally (bonding surface perpendicular to dentinal tubules) or axially (parallel to tubules) in an acrylic base. An orthodontic grinder was used to remove the enamel, while paying special attention to ensure that pulp exposure did not occur. The speci-

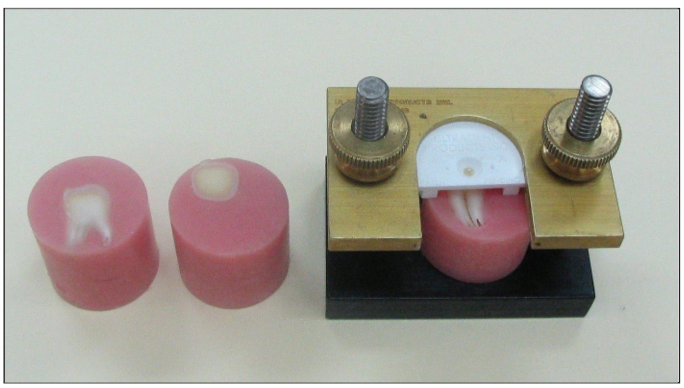

Figure 1. *Ultradent mold setup for the shear bond strength test (left: axial dentin surface; middle: occlusal dentin surface; right: Ultradent jig setup on the axial dentin surface).*

mens were then polished on moist 600-grit sandpaper for 30 seconds in a randomized figure-eight pattern, rinsed and stored in 37°C water. Any specimen with a pulp exposure was immediately excluded from the experiment. Modified pH solutions were made by combining the correct volume of each component of the Experimental Self-Etch adhesive (ExpSE). Solid potassium hydroxide was added with agitation, and the pH was determined using a calibrated Schott pH electrode. Solid potassium hydroxide was added until the desired pH was obtained. Immediately after the pH was measured, the solution was used to bond either axially- or occlusally-mounted dentin specimens. Application of the adhesive consisted of two consecutive coats with 10 seconds of agitation, air drying until no adhesive movement was observed on the dentin surface, followed by a light cure for 10 seconds at 500 mW/cm^2 (VIP, BISCO, Inc). Aelite All-Purpose Body translucent composite (BISCO, Inc) was used to fabricate the posts using an Ultradent mold (Ultradent Products, Inc, South Jordan, UT, USA) (bonding area 4.45 mm^2) (Figure 1) and light-cured from the top for 40 seconds. The mold was removed and the specimens stored in water at 37°C for two hours. The specimens were then tested until failure on a Universal Testing Machine (Instron, Model 4466; Canton, MA, USA) using a crosshead speed of 1 mm/minute. The Instron readings (kg) were then converted to bond strength in MPa.

Table 1: *Shear Bond Strength (SBS) on Unetched Dentin with Different Tubule Orientations*

pH of Adhesive	SBS, MPa, Axial	SBS, MPa, Occlusal
1.1	29.8 (6.6)^{a,1,*}	11.5 (3.0)^{b,2}
1.8	28.0 (3.8)^{a,1}	14.5 (5.7)^{b,2}
2.3	26.7 (4.6)^{a,1}	22.6 (3.9)^{a,1}
2.7	27.0 (6.3)^{a,1}	23.2 (3.0)^{a,1}

*Standard deviations are shown in parentheses (axial: N=10; occlusal: N=6). Values with the same superscript letters (a,b) in the same column or same superscript number (1,2) in the same row are not statistically different (p>0.05). Groups with * had one specimen with a debonding dentin fracture, and the other groups had no observed fractures of dentin or composite post (via visual exam).*

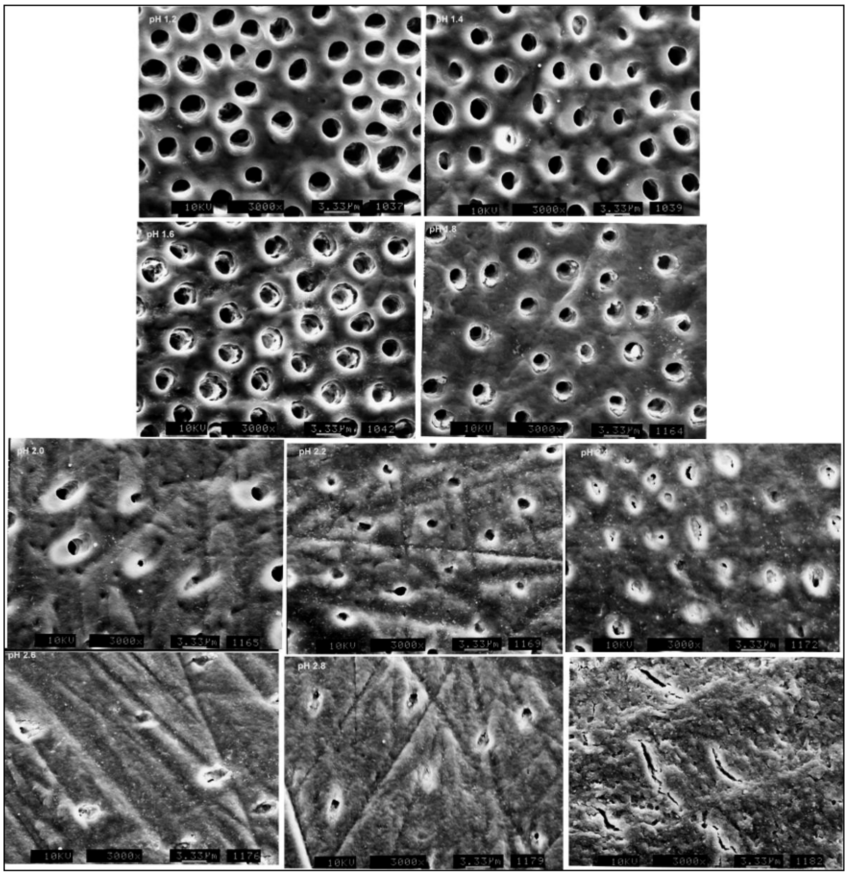

Figure 2. *SEM pictures of the occlusal dentin surface after 15-second etching using a phosphoric acid solution (32 wt%) with an adjusted pH of 1.2, 1.4, 1.6, 1.8, 2.0, 2.2, 2.4, 2.6, 2.8 and 3.0 (magnification 3,000x).*

The morphology of the etched occlusal mid-dentin surface was examined by Scanning Electron Microscope (SEM) (Topcon, Model SM-510; Tokyo, Japan). The pH of aqueous phosphoric acid solutions (32 wt%) was adjusted to 1.2, 1.4, 1.6, 1.8, 2.0, 2.2, 2.4, 2.6, 2.8 and 3.0 by adding potassium hydroxide. Occlusal portions of the tooth specimens were ground off to expose flat dentin surfaces polished on moist 600-grit sandpaper for 30 seconds in a randomized figure-eight pattern and rinsed with water. The polished dentin was then etched for 15 seconds using a phosphoric acid solution with an adjusted pH. The phosphoric acid solution was rinsed with water. The etched dentin surface was then subjected to SEM examination. The opened dentin tubule size was measured and the total area of open tubules was calculated.

All data analysis was done by ANOVA, followed by a Student-Newman-Keuls multiple-range test.

RESULTS

The results reported in Table 1 are shown with indications of which groups were statistically different ($p<0.05$), as indicated by the superscript letters or numbers next to the results.

Determination of the shear bond strength (SBS) using the method developed by Ultradent showed that, when bonding to axial dentin, bond strength is not dependent on the pH of the self-etch adhesive (pH>0.05). However, when bonding to occlusal dentin, the bond strength is significantly affected by the pH of the self-etch adhesive

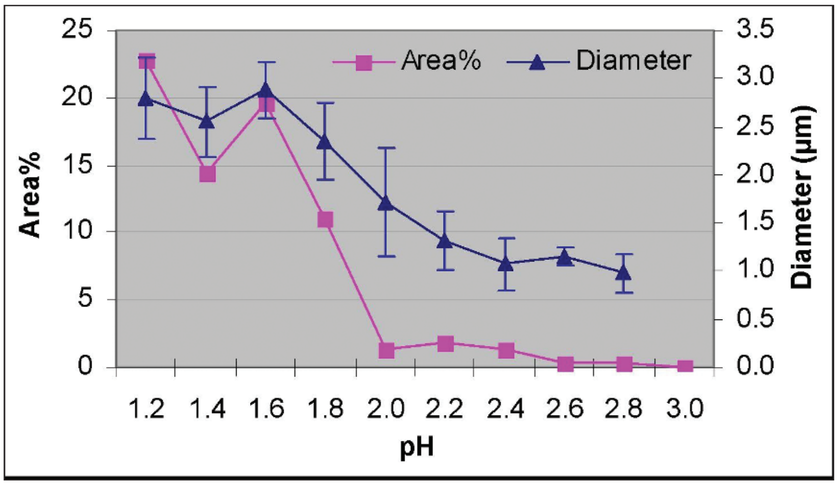

Figure 3. *The diameters (μm) of opened dentin tubules and the percentage (%) of dentin surface area occupied by opened t ubules in relation to the pH of etchant.*

($p<0.05$). With a pH higher than 2.3, the bond strength showed no difference ($p>0.05$) between the occlusal and axial dentin, while with a pH lower than 1.8, the tubule orientation (occlusal or axial) had a significant effect on the bond strength.

SEM pictures of the etched occlusal dentin surfaces are shown in Figure 2. This figure shows that the smear layers and smear plugs were completely solubilized by phosphoric acid solution with a pH adjusted to 1.2 and 1.4. At a higher pH, the smear layers and smear plugs were partially solubilized. At pH 3.0, the smear layer/plug was completely retained. The diameters (μm) of opened dentin tubules and the percentage (%) of the dentin surface area occupied by opened tubules in relation to the pH of etchant are shown in Figure 3.

DISCUSSION

Dentin adhesion is a synergistic process that includes physical, chemical and/or mechanical bonding components.[23] Physical bonding forces, such as van der Waal forces or hydrogen bonding, are always present but weak. Chemical bonding is strong, including covalent, ionic, chelation or metallic bonding. Mechanical bonding/interlocking is the most effective and common means of creating strong bonds.

To investigate the effect (acidity) a self-etching system plays on the resulting smear layers/plugs, the authors of the current study conducted a SEM evaluation of occlusally-etched dentin surfaces using pre-adjusted pH solutions. The SEM examinations showed that, at a pH of 1.2 and 1.4, the smear layer and plugs were completely removed. The mean tubule diameter was 2.6 ~ 2.8 μm, which is similar to the size of a fully opened dentin tubule close to the pulp, as reported in the literature (diameter 2.4 ~ 3.0 μm).[25] The area of opened dentinal tubules occupied about 15%-22% of the total dentin surface area. The SEM results clearly showed that the dentin tubules were fully opened and the smear layers and plugs were completely dissolved by etchant at this low pH. At pH 1.6, the smear layers were completely removed, while the smear plugs were partially retained. The diameter of the opened tubule was approximately 2.9 μm, and the area of the opened tubules occupied about 20% of the total dentin surface. At pH 1.8, significantly less dentin tubules were opened (11% of the total area), and the size of the opened tubule had significantly decreased (2.3 μm in diameter). At pH 2.0-3.0, a negligible amount of smear layers and smear plugs were dissolved (opened tubules occupied less than 2% of the total dentin surface area). The smear layers and plugs were completely retained at pH 3.0.

Self-etch adhesives can be divided into "strong/aggressive" (pH < 1), "intermediately strong/aggressive" (pH ≈ 1.5), "mild" (pH ≈ 2) and "ultra-mild" (pH ≥ 2.5) adhesives, depending on their self-etching and demineralization capability.[24] In self-etch systems, the aggressive adhesive is able to partially or completely dissolve the smear layers and plugs and open the dentin tubules.[22] With the bonding surface perpendicular to the tubule (occlusal bonding), small resin tags might be formed to provide mechanical interlocking for dentin bonding. However, the results of the current study showed that bond strength significantly decreased on occlusal dentin with a low-pH (pH<1.8) self-etch adhesive. (The visual exam of the debonding surface showed there was one dentin fracture for the Axial-pH1.1 group, while the other groups had no observed fractures of dentin or composites. It would be more valuable to use SEM for investigating the debonding failure modes).

It is speculated that the occlusal orientation of dentin tubules causes water that is present in the dentin tubules to be more available to migrate to and/or through the cured adhesive. This is affected by the pH, since a lower pH would lead to removal of smear debris that would potentially block the water in the dentin tubules. The migrated dentinal fluid can result in increased dentin surface wetness, which can interfere with adhesion between the adhesives and dentin, since the relatively hydrophobic monomers in adhesives do not adhere to a highly hydrophilic dentin surface. In addition, the dentinal fluid might also diffuse through the cured adhesive layer and cause a lack of adhesion (plasticizing) of the overlying composite or resin cement. This is similar to the explanation put forth by Tay and others, as evidenced by lower bond strength, water bubbles and water tree formation found in their studies.[17-19] Water in the polymer matrix (cured adhesives) exists in three states: hydrogen-bonded, partially hydrogen-bonded, and free water. Free water forms a water channel, creating a pathway in the polymer matrix for the fast diffusion of water. The polymer matrix of cured self-etch adhesives is extremely thin (5-20 microns) and hydrophilic, providing perfect conditions to act as a permeable membrane on top of the wet dentin.

Tay and others reported a study where a light-cure composite was placed on top of cured acidic single-step adhesives, such as One-Up Bond (Tokuyama Dental Corporation, Taitou-ku, Tokyo, Japan) and Prompt L-Pop (3M ESPE). The composites were light-cured immediately or with a time delay of 2.5, 5.0, 10 and 20 minutes. The purpose of the delayed light-activation was to intentionally allow the possible diffusion of dentinal fluid through the cured adhesives. The study showed that significantly lower bond strengths were found upon prolonged light-cure delay (5, 10 and 20 minutes).[26] In contrast, there was no difference shown with 20-minute delay light activation for the composite placed on a hydrophobic and less permeable resin (D/E resin, BISCO, Inc).[26] When the same delayed curing of composites was done on dehydrated dentin, the bond strengths were almost the same as those of immediate curing. This provided compelling evidence that the migration of dentinal fluid through the adhesive layer significantly interfered with the bond between the adhesives and composites.

These results may lead to speculation as to the importance of the effect of micro-mechanical interaction in bonding (even if resin tags are formed with removal of the smear layers/plugs, the migrated dentinal fluid can affect the strength of the dentin-adhesive interface[4]). Finally, it is worth noting that the smear layers and smear plugs are shown to be acting as a barrier of dentinal fluid and limit fluid penetration within the resin dentin interface. Low pH self-etching adhesives (strong/aggressive) might be desired for better etching of enamel, but they are shown to remove the smear layers and smear plugs, increase smear layer permeability, increase dentinal fluid flow under pressure and cause interference in the adhesive interface, resulting in significantly lower initial bond strengths.

CONCLUSIONS

In self-etch systems, the pH of adhesives did not have any effect on shear bond strength when bonding parallel to the tubule (axially-oriented dentin). However, a lower pH caused lower bond strengths when bonding perpendicular to the tubule (occlusally-oriented dentin). With a pH of less than 1.8, bond strengths on occlusal dentin were significantly lower. The clinical implication is that the results reported for bond strengths should be understood and compared based on the orientation of the dentin used for the measurements, particularly for those adhesives with a very acidic system (pH lower than 2). This may also explain some of the differences reported by the different laboratories for the same materials, which, in the past, has been hard to reconcile.

(Accepted 9 October 2010)

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