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ORIGINAL ARTICLE

Multiple Coatings can Improve the Bond Durability of One-step Self-etching Adhesive to Primary Dentin

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Objective To investigate whether multiple coatings can improve the bond durability of one-step self-etching adhesive to primary dentin.

Methods Twelve caries-free human primary molars were randomly divided into 2 groups. In group 1, each tooth was hemisected into 2 halves. One half was assigned to the control subgroup 1, which was bonded with a commercially available one-step self-etching adhesive according to the manufacturer's instructions; the other half was assigned to experimental subgroup 1, in which the adhesive was applied three times before light curing. In group 2, one split half tooth was bonded with a commercially available one-step self-etching adhesive according to the manufacturer's instructions; for the other half, three layers of adhesive were applied with each successive layer of light curing. Specimens were stored in 0.9% NaCl containing 0.02% sodium azide at 37°C for 18 months and then were subjected to microtensile bond strength test and the fracture mode analysis.

Results When the adhesive was applied three times before light curing, the bond strength of the experimental subgroup 1 was significantly higher than that of the control subgroup 1 (47.46 ± 13.91 vs. 38.12 ± 11.21 MPa, $P < 0.05$). When using the technique of applying multiple layers of adhesive with each successive layer of light curing, no difference was observed in bond strength between the control subgroup and the experimental subgroup (39.40 ± 8.87 vs. 40.87 ± 9.33 MPa, $P > 0.05$).

Conclusion Multiple coatings can improve the bond durability of one-step self-etching adhesive to primary dentin when using the technique of light-curing after applying 3 layers of adhesive.

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ONE-STEP self-etching adhesives were developed in order to shorten the bonding procedure and reduce the sensitivity of the technique, for all of the components are blended in one solution. However, some previous studies have identified some concerns about the performance of these adhesives such as reduced dentin bond strength and poor bond durability.^{1,2} Several possible reasons are responsible for the poor bonding performance of these simplified adhesives: (1) the presence of highly hydrophilic monomers that are sensitive to water sorption from the underlying dentin,³ increasing hybrid layer permeability and nano-leakage;⁴ (2) a differential infiltration gradient through the dentin related to differences in the molecular weight of the adhesive system compounds;⁵ (3) a high concentration of protic solvents and dissolved molecular oxygen within the adhesive layer contributing to poor evaporation;⁶ (4) the limited thickness of the adhesive layer, which may also magnify the oxygen inhibition effect on adhesive polymerization, and (5) their low degree of cure.⁷

Interactions of the self-etch agents with dentin are limited by many factors from both the substrate and the adhesive itself. Optimal resin infiltration is important to achieving adequate bond strengths, as the effective resin-dentin bonds may only be formed if primers and resins are able to penetrate through the smear layer and interact with the underlying dentin.⁸ The infiltration of the adhesive blend, solvent/water evaporation, and thickness of the adhesive layer are directly related to chemical and rheological properties,⁶ but may be influenced by the application mode.⁹ It has been hypothesized that more uniform adhesive infiltration and greater solvent/water evaporation would be achieved when bonding with multiple coatings⁷ or by a prolonged application time of the primers/adhesives onto the dentin surface.¹⁰

Although there have been studies on the effects of multiple coatings on bond strength of resin to permanent dentin,¹¹⁻¹⁵ so far as we know, there is few studies to evaluate the effects of multiple coatings on the bond strength to primary dentin.¹⁶ In one of our previous studies, the effect of multiple coatings of a one-step self-etching adhesive system on the immediate bond strength to primary dentin was evaluated.¹⁷ This study was to further assess the effect of multiple coatings on the bond durability of a one-step self-etching adhesive to primary dentin.

MATERIALS AND METHODS

Tooth preparation

Twelve caries-free second primary molars, which had been

extracted because of prolonged retention, were collected after obtaining the informed consent of children's parents. The study protocol was reviewed and approved by the Ethics Committee for Human Studies, Peking University, Beijing, China. The teeth were stored in 0.9% NaCl containing 0.02% sodium azide at 4°C for no more than one month.

The occlusal enamel of each tooth was removed. The teeth were hand polished on a wet 600-grit silicon carbide paper for 30 seconds to create a realistic smear layer on the surface of the occlusal mid-coronal dentin.

Bonding procedures

The 12 prepared teeth specimens were randomly divided into 2 groups, 6 teeth in each group.

In group 1, each tooth was hemisected into 2 halves using a low-speed water-cooled saw equipped with a diamond-impregnated disk (Isomet 1000; Buehler Ltd., Lake Bluff, IL, USA). One half tooth assigned to control (C1) subgroup was bonded with a one-step self-etching adhesive (Clearfil S3 Bond, lot no. 00143A; Kuraray Medical, Osaka, Japan) according to the manufacturer's instructions. The composition of the one-step self-etching adhesive is shown as follows: 10-methacryloyloxydecyl dihydrogen phosphate (MDP), bis-phenol A diglycidylmethacrylate (Bis-GMA), 2-hydroxyethyl methacrylate (HEMA), hydrophobic dimethacrylate, camphorquinon, ethyl alcohol, silanated colloidal silica, and water. After undisturbed for 20 seconds, the adhesive was uniformly spread by a strong air stream for 10 seconds and then light-cured for 10 seconds using a halogen light-curing unit with an output of 700 mW/cm². The other half tooth assigned to experimental (E1) subgroup was continuously bonded with the adhesive for 3 times prior to light-curing for 10 seconds. After an 8-mm-height composite resin block (Clearfil AP-X, Kuraray, Osaka, Japan) was built up on the bonded surfaces by 5 or 6 sequential increments of resin, each one was light-cured for 20 seconds.

In group 2, the dentin surface of one split half (C2 subgroup) was bonded with the one-step self-etching adhesive according to the manufacturer's instructions. For the other half (E2 subgroup), adhesive was applied to the dentin surface. After leaving undisturbed for 20 seconds, the adhesive was spread by a strong air stream for 10 seconds and light-cured for 10 seconds. This procedure was repeated for 3 times, followed by composite buildups.

Microtensile bond strength test

After stored in distilled water at 37°C for a week, the bonded teeth were longitudinally sectioned across the

bonded interface into sections with a diamond saw (Isomet 1000), to produce a series of 0.8 mm×1.5 mm×8.0 mm beams. Four to six beams were obtained from each specimen. These beams were then trimmed to form hourglass shape specimen with a cross-sectional area of approximate 0.6 mm². Then specimens were stored in 0.9% NaCl containing 0.02% sodium azide at 37°C for 18 months before bond strength test. The storage solution was changed each month.

After 18 months of water storage, the specimens were individually fixed to a custom-made testing jig with cyanoacrylate glue (Universal Instant Adhesive, Henkel Adhesives Co. Ltd., Shantou, China). The specimen was then subjected to tensile load at a crosshead speed of 1.0 mm per minute until failure (Autograph DCS-5000, Shimadzu, Japan). The bond strength values were recorded as Newton (N) and later converted to megapascals by dividing the loads in Newtons by the total bonded surface area (MPa= N/mm²).

Debond pathway determination

Both surfaces of each fractured specimen were observed under a stereomicroscope (Olympus 220670, Tokyo, Japan) with 40× magnification to record the failure modes. The fracture modes were classified as follows: (1) cohesive failures in the composite resin were defined as the fracture occurred exclusively within the resin composite; (2) failures in the adhesive joint were confirmed if the fracture site was entirely within the adhesive; (3) cohesive failures in dentin were the fracture occurring exclusively within the dentin; and (4) mixed failures were verified if the fracture site continued from the adhesive into either the resin composite or dentin.

Statistical analysis

We tested all data using SPSS statistical software (version 16.0). The microtensile bond strength was expressed as mean±SD. Two-way analysis of variance (ANOVA) and *post hoc Tukey* tests were used to compare the effects of the dentin treatments on bond strength. Monte Carlo method was used to analyze the distribution of the fracture

mode. The statistical unit was beams, not teeth. Statistical significance was pre-set at $\alpha = 0.05$.

RESULTS

Of a total of 123 specimens, only 4 specimens failed, and 1 was from subgroup C1, 1 from subgroup E1, and 2 from subgroup E2. All failure occurred during specimen preparation for bond strength test. Finally, the sample size of C1, E1, C2 and E2 subgroups was 31, 30, 28, and 30, respectively.

Comparison of bond strength

In group 1, the bond strength of the E1 subgroup was significantly higher than that of the C1 subgroup (47.46±13.91 vs. 38.12±11.21 MPa, $P<0.05$). In group 2, there was no difference in bond strength between the C2 and E2 subgroups (39.40±8.87 vs. 40.87±9.33 MPa, $P>0.05$).

Distribution of the fracture mode

Table 1 summarizes the distribution of the fracture modes. Adhesive joint failures were the most common fracture pattern observed in both the control and experimental subgroups. The incidence of cohesive failure in the composite resin in E1 subgroup was significantly higher than that in the other three subgroups (all $P<0.05$).

DISCUSSION

As shown in this study, after 18 months of water storage, the bond strength of the E1 subgroup was higher than all the other three subgroups, indicating that applying 3 layers of adhesive before light curing can improve the bond durability of the one-step self-etching adhesive to primary dentin. No significant difference was observed between the C2 subgroup and E2 subgroup, indicating that the technique of applying 3 layers of adhesive with curing each successive layer can not improve the bond durability of the one-step self-etching adhesive to primary dentin. However, compared with the immediate bond strength

Table 1. Distribution of the fracture mode of the four study groups

Groups	Sample size	Fracture mode [n (%)]			
		Cohesive failures in the composite resin	Failures in the adhesive joint	Cohesive failures in dentin	Mixed failures
Group 1 C1 subgroup	31	3 (9.68%*)	21 (67.74%)	3 (9.68%)	4 (12.90%)
E1 subgroup	30	10 (33.33%)	14 (46.67%)	3 (10.00%)	3 (10.00%)
Group 2 C2 subgroup	28	4 (14.29%*)	20 (71.43%)	1 (3.57%)	3 (10.71%)
E2 subgroup	30	3 (10.00%*)	21 (70.00%)	3 (10.00%)	3 (10.00%)

* $P<0.05$ compared with E1 subgroup.

reported in our previous study,¹⁷ the bond strength decreased in all subgroups after 18 months water storage, indicating that the technique of multiple coatings can not totally prevent the bond aging of the one-step self-etching adhesive.

There are minor differences between primary and permanent teeth in terms of dentin composition and morphology.^{18, 19} It has been pointed out that the concentration and diameter of dentinal tubules is higher in primary dentin than that in permanent dentin. The amount of surface moisture was also found to be higher in primary dentin.^{20, 21} All these factors alter the effectiveness of the dentin conditioners, hence affecting the bond strength.^{20, 21} However, people often transfer the knowledge about permanent teeth to primary teeth, regardless of these microstructural differences between permanent and primary dentin. Therefore, the value of the studies on permanent teeth, such as the technique of multiple coatings which might alter the conditions of bond surface and the quality of the hybrid layers, is worthy to be ascertained on primary teeth.

Although the effects of multiple coatings on the immediate bond strength were well investigated, there are only a few studies evaluating the effect of multiple coatings on the bond durability of dental adhesives to permanent teeth and to primary teeth. Reis *et al*²² evaluated the immediate and 6-month resin-dentin bond strength of one-step self-etch systems under double application mode. They concluded that double application of two one-step self-etch adhesive (Xeno III and iBond) could improve the bond durability. Similar results were obtained in this study, although different adhesive and different adhesive layers were used. For the teeth bonded with one-step self-etching adhesives, penetration of the adhesive into dentin occurs simultaneously with demineralization of the dentin. When the resin fails to completely infiltrate the deeper portions of the demineralized dentin, the bond between resin and dentin might be weakened.²³ Applying multiple consecutive coatings prolongs adhesive application time, which might allow resin monomer to penetrate into the total depth of the demineralized dentin.^{24, 25} Besides, Clearfil S3 Bond contains a significant amount of water and solvent which are expected to be removed by air blowing after application of the adhesive.¹¹ Multiple coatings also prolong air-drying time, therefore can remove residual water and solvent more effectively.¹³ This has been confirmed that the prolonged application and subsequent solvent evaporation may improve resin infiltration within the exposed collagen fibers.^{13, 26} Therefore, the use of multiple layers of adhesive before curing, as done in this study, allows more time for

adhesive diffusion and removal of residual water, thereby, improving resin infiltration and crosslinking of the adhesive comonomers within the hybrid layer.

When each successive layer is light cured, the adhesive layer becomes thicker without changing the quality of the hybrid layer, which can create a relatively thick intermediate layer with low elastic modulus between dentin and composite. It has been speculated that as the component overlying the hybrid layer, the adhesive layer might help to preserve the integrity of the hybridized dentin, protect it from polymerization shrinkage stresses, act as a stress absorbing layer, and contribute to improving and maintaining bond strength.^{27, 28} However, each adhesive system has an ideal thickness to reduce interface stress while preserving the interface integrity. And quite different results were achieved in different studies: D'Arcangelo *et al*¹² reported an excess of adhesive layer thickness could negatively influence the strength. Pashley *et al*⁷ and de Silva *et al*²⁹ demonstrated an increased bond strength using the technique of applying multiple layers of adhesive then curing each successive layer. This is probably due to the differences in procedures, application modalities, and compositions (solvent agents and filling materials) of the various commercially available bonding systems used.³⁰ It is worth emphasizing that, to date, no study has successfully established a positive correlation between the thickness of the resin-infiltrated layer and bond strength, suggesting that the quality, rather than the thickness of the resin-infiltrated layer, is more important for bond strength measurements.^{27, 31}

Analyzing the failure modes can help to compare the bond strength. Microtensile bond strength test is considered the most credible method for evaluating dentin adhesion. With using small sized specimens (1 mm²), it offers several advantages including optimal stress distribution at the resin/dentin interface, more adhesive failures (that is, true bond strength values), fewer cohesive failures, and a higher interface bond strength.³² In the E1 subgroup, the incidence of cohesive failures in the composite resin was significantly higher than that in the other three subgroups. This result indicated higher bond strength in the E1 subgroup.

In conclusion, applying 3 layers of one-step self-etching adhesive before light curing might be able to improve the bond durability of resin to primary dentin.

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