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ORIGINAL

The effects of different application time on all-in-one self-etching adhesives

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ABSTRACT : The purpose of this study was to evaluate the effects of different application times in all-in-one systems on micro-tensile bond strength. Three all-in-one systems, EB (Easy Bond, 3M ESPE, St. Paul, MN, USA), BB (BeautiBond, Shofu Inc., Japan) and BF (BondForce, Tokuyama Denetal, Ibaraki, Japan) were used with four different application times: 10s, 20s, 30s, and 40s. Bond strengths to dentin were determined using μTBS test after 24h of water storage at 37℃. In addition, an evaluation of both resin-dentin interface, and fractured surface on dentin side was performed by SEM. The maximum μTBS for each system was observed with a 20s application time. There was no positive correlation between the thickness of hybrid layers and application time, which meant hybrid layers did not increase in thickness along with the increase in application time, rather a difference of quality was detected. Prolonged application time affected the adhesive strength of all-in-one systems, and they were mostly effective for the 20s application time.

Key words : Microtensile bond strength, All-in-one adhesive, Failure modes, Application time

INTRODUCTION

With the rising popularity of the conception of minimal invasive dentistry[1] and the increasing esthetic demand from patients, resin adhesive system has gradually become the mainstream of dental restoration, and bonding technology is a key factor of this system. The introduction of acid etching by Dr. Michael Buonocore[2] has a significant impact on the blooming of the adhesive dentistry. Since tooth-bonding techniques have various clinical applications, the research of improving bond strength and simplifying the bonding procedure has attracted a lot of attention to many researchers.

According to smear layer removal methods and different formation process of "hybrid layer", there are two main categories of dental bonding systems, which are total-etch adhesive system and self-etch adhesive system. Total-etch adhesive system has a separate phosphoric acid etching step and it can entirely remove the smear layer. On the other hand, self-etch adhesive system relies on acidic monomers to dissolve the smear layer and demineralize the tooth structure. Self-etch adhesives have lower acidity, and with self-etch adhesives, the depth of demineralization and resin infiltration is the same. They are easy to use, and have low technique sensitivity and low postoperative sensitivity[3-7]. So it is welcomed by many clinicians, accepted the self-etch adhesives especially the all-in-one system.

The clinicians and the researchers expect the simplification of operational procedure and the improvement of dental prosthesis retention. Bonding strength depends on the extent of penetration of the demineralized dentin and form a high-quality hybrid layer. It proved that the hybrid layer will increase the spread and wettability of dental self-etching primer[8]. The interaction effects between self-etch adhesive and hybrid layer may be associated with the kinds of self-etch adhesive. Thus in the self-etch adhesive system, the buffer effect of application time plays a necessary role during the operation and will finally determine the quality of dental hybrid layer[9].

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For this reason, while improving fillers, solvents, and primers for dental adhesives, researchers now are paying more attention to each step of clinical bonding procedure, such as surface treatments, time of treatment, coating methods, and application time. So far, however, there is no explicit guidance about the application time in the use of all-in-one self-etch adhesives. One way to improve the demineralization effect and consequent hybrid layer quality is to increase acidic monomer concentration by prolonging the application time[10-12]. Some studies showed that the acidly of monomer and its prolong action have the potential to enhance the infiltration of resin resulting in a highly cross-linked hybrid layer[11, 12].

The purpose of this study therefore is to evaluate the effect of application time of all-in-one systems on micro-tensile bond strength after 24-hour storage. The null hypothesis is that there is no effect on bond strength when using different application time in all-in-one systems.

MATERIALS AND METHODS

Teeth used

72 extracted non-carious human molars were used in this study to test three different all-in-one systems. Each system consisted of 24 teeth which were further divided into 4 groups with 6 teeth in each group. The teeth were collected under a protocol reviewed and approved by the institutional review board of Hokkaido University. The teeth were stored at 4℃ in an aqueous solution of 0.5% Chloramine-T, and used within four months after extraction. Flat dentin surfaces were obtained by removing the coronal enamel of each tooth in a gypsum model trimmer with the water coolant, leaving the surrounding enamel, making sure that the remaining dentin thickness was 2.5 ± 0.2 mm, as measured with a caliper. Dentin surfaces were then ground with 600-grit SiC paper for 60 s under continuous water-cooling to produce a standardized smear layer prior to bonding.

Adhesives

Three commercially available all-in-one adhesives were employed in this experiment: Easy Bond (EB, 3M ESPE, St. Paul, MN, USA), BeautiBond (BB, Shofu Inc., Kyoto, Japan), and BondForce (BF, Tokuyama Dental, Ibaraki, Japan). Table 1 shows the chemical formulations and the respective manufacturer's instructions for usage of these three adhesives.

In accordance to the application time each adhesive was randomly assigned to four groups, that is, 10 sec group, 20 sec group, 30 sec and 40 sec group. All bonded surfaces were build-up with resin composite (Clearfil AP-X, Kuraray Medical Inc.; Okayama, Japan, Shade A3, Lots: 01320A) in increments to a thickness of 5 mm. Each incremental layer was light cured for 20 s (light output intensity properly controlled at no less than 550 mW/cm2). The adhesive specimens were stored in distilled water at 37℃ for 24 h.

Table 1 Chemical formulation and the respective manufacturer's instructions

Materials (Lot No.)	Ingredients	Direction for use
Easybond (364117) (3M ESPE, St Paul, MN, USA)	HEMA, Bis-GMA, Methacrylated phosphoric esters, 1,6 hexanediol dimethacrylate Methacrylate functionalized Polyalkenoic acid (Vitrebond™ Copolymer) Finely dispersed bonded silica filler with 7 nm primary particle size, Ethanol, Water	1 apply bonding for 20s 2 blow gently without scattering for 5s 3 light-cure for 10s
BeautiBond (120815) (SHOFU Inc., Kyoto, Japan)	Acetone, Bis-GMA, TEGDMA, 4MET, 6MHPAc, pure water and others.	1 leave undisturbed for 10s after the end of application 2 blow gently without scattering for 3s and then blow more strongly 3 light-cure for 10s
BondForce (078069A) (Tokuyama Dental, Ibaraki, Japan)	HEMA, Bis-GMA, TEGDMA, alcohol, phosphoric acid	1 leave undisturbed for 20s after the end of application 2 blow gently without scattering for 5s and then blow strongly for 5s 3 light-cure more than 10s

Bis-GMA: bis-phenol A diglycidylmethcrylate; HEMA: 2-hydroxyrthl methacrylate; MDP: 10-methacryloyloxy decyl-dihydrogen phosphate; TEGDMA: triethylene glycol dimethacrylate;

Micro-tensile Bond Strength (μTBS) Test

After 24 h water storage in 37℃, 6 resin-bonded sticks (lmm X lmm approximately) were obtained from each tooth using a diamond saw (Isomet Low Speed Saw, Buehler, Lake Bluff, IL, USA). The sticks were then fixed to a Ciucchi's jig with cyanoacrylate glue (Model Repair II Blue, Dentsply-Sankin, Otahara, Japan) and subjected to a tensile force at a crosshead speed of 1 mm/min in a desktop testing apparatus (EZ test, Shimadzu, Kyoto, Japan) until failure occurred. The μTBS was expressed in MPa, dividing the applied force (N) at the time of fracture by the bonded area (mm^2). To compare the groups formed by the adhesive with the different application time, one-way ANOVA and Games-Howell test were used at $p < 0.05$.

Failure modes

The modes of failure were determined using a light-microscope (x20, Magnifier Light, Asone, Osaka, Japan). The failure modes were categorized as:

Mixed with dentin cohesive failure: adhesive + cohesive in dentin.

Adhesive: adhesive failure at the resin-dentin interface only.

Cohesive: adhesive + cohesive in resin composite

SEM observation

1) Observation of the dentin surface after μTBS testing

The fractured surface of the dentin-side from all specimens after the μTBS test were dried overnight in desicators at room temperature, then sputter-coated and observed using FE-SEM (S-4000, Hitachi, Tokyo, Japan) with an accelerating voltage of 10 kV.

2) SEM observation of the resin-dentin interface

To observe the morphology of the resin-dentin interface, the newly prepared resin-bonded specimens were sectioned perpendicularly to the adhesive interface, using an Isomet saw, to obtain two slabs of 2 mm thickness of each system. The cut surfaces were sequentially polished with 600-, 800-, and 1000-grit silicon carbide papers under running water. This was followed by polishing sequentially with 6-, 3-, 1-μm diamond pastes (DP-Paste, Struers, Denmark), and cleaning with an ultrasonic device between each diamond paste polish. After polishing, the specimens were immersed in 1 M hydrochloric acid for 30 s and 5% sodium hypochlorite for 5 min, followed by rinsing with water. After drying, the specimens were sputter-coated with Pt-Pd for 120 s. The resin-dentin interfaces were then observed using a scanning electron microscope (SEM, S-4000, Hitachi, Tokyo, Japan).

RESULTS

Micro-tensile Bond Strength (μTBS) Test

The bond strength of all three adhesives all reached the maximum value when prolonged to 20s, but it declined greatly when application time was 40s. In the comparison between 10s and 20s application time, there was no significant difference in bond strength ($p>0.05$). But the comparison between 20s and 30s application time group showed significant differences in bond strength ($p<0.05$), and the bond strength began to decrease. There was no significant difference between the 10s and 30s application time. In the groups of EB and BB, the application time of 30s and 40s had significant differences. But in the groups of BF, the application times of 10s, 30s and 40s did not have statistically significant differences between each other.

Failure Analysis

In fig1 and fig2, according to the rank sum test of K independent Samples with different failure patterns, it showed that different adhesive failure patterns did not have statistically significant differences ($p>0.05$).

SEM observation of fracture surface

In fig3, the application time of 10s and 20s had better sealing abilities of dentinal tubules than that of 30s and 40s.

Among the groups of EB, the dentinal tubules were greater in diameter and the group had a more regular pattern in the application of 20s. The dentinal tubules of 10s were thinner and irregular, but had a good sealing condition. The tubular diameter became smaller and the pattern became irregular when the application time of 30s and 40s were applied. Some dentinal tubules of 30s and 40s group lacked resin tags and their sealing ability were worse than those in the previous groups with 10s and 20s application time.

Among the groups of BB, the dentinal tubules with the application of 10s and 20s had a regular pattern and their resin tags were equally sealed. In the application group of 30s, some dentinal tubules lacked tags and sealing ability is worse than those of 10s and 20s. When adhesives were applied for 40s, sand-like substance appeared, and began to cover the top of the dentinal tubules. At the same

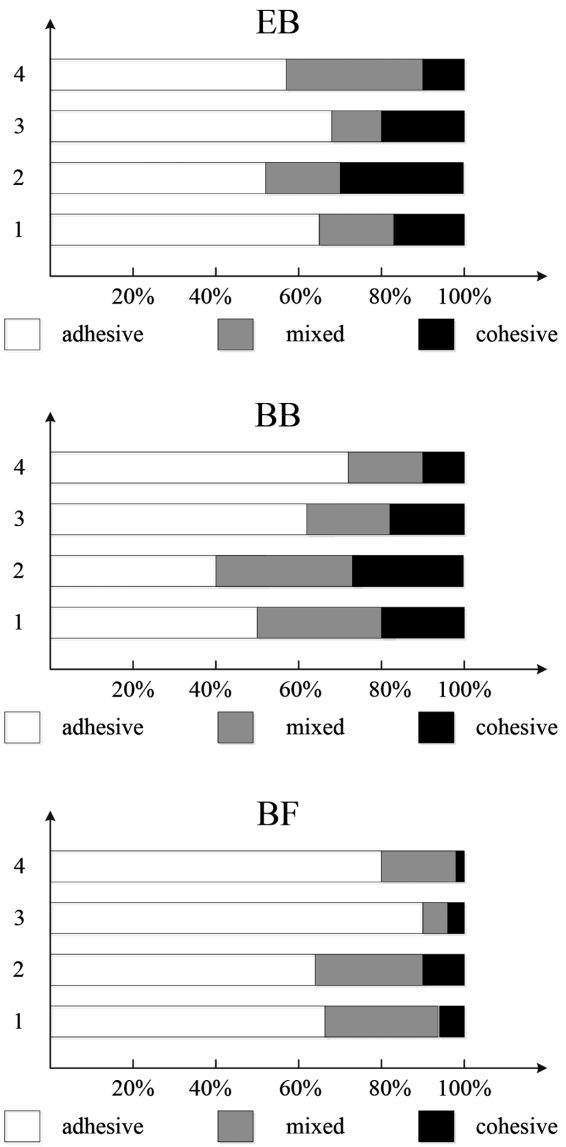

Fig. 1 The percentage of fracture modes (n=30/group). mixed: adhesive+cohesive in dentin.

adhesive failure at the resin-dentin interface only. cohesive: adhesive+cohesive in resin composite. For the three adhesives, the failure mode had a tendency to show the higher incidence of adhesive failure (at the resin-dentin interface only) at the optimal duration.

time, some of the tubules lacked resin tags.

Among the groups of BF, the sealing ability and the filling of dentinal tubules are satisfied only in the 20s application group. There were obvious appearance of sand-like material on the top of the tubules and some of which also lack resin tags in the application of 10s, 30s and 40s application groups.

SEM observation of the resin-dentin interface

In fig4, no difference could be found in the thickness of hybrid layers formed by the tested adhesives. All three self-etch adhesives formed resin tags, which were usually long and thin.

Among the groups of EB, the thickness of adhesive layers increased along with the increase of application time. The continuous dense resin tags were observed in 10s and 20s group. The diameter and the shapes of the resin tags were regular. However, in the 30s and 40s groups, the number and the continuity of resin tags decreased and the shapes and diameter became irregular.

In the groups of BB, the thickness of adhesive layers also increased along with the increase of application time. Resin tags with the application time of 20s, were dense, regular in shape and without voids. When adhesives were applied for 10s, some of the resin tags were fractured and their continuity decreased. When adhesives were applied for 30s, the density of resin tags declined. In the groups of 40s groups, the resin tags were short and their shapes and diameters were irregular.

Among the groups of BF, the thickness of adhesive layers also increased along with the increase of application time. When adhesives were applied for 20s, the resin tags were dense but some of them were fractured. In the 10s group, fewer resin tags were formed. These resin tags were irregular in shape and very fragile. The similar phenomenon was observed in 30s and 40s groups.

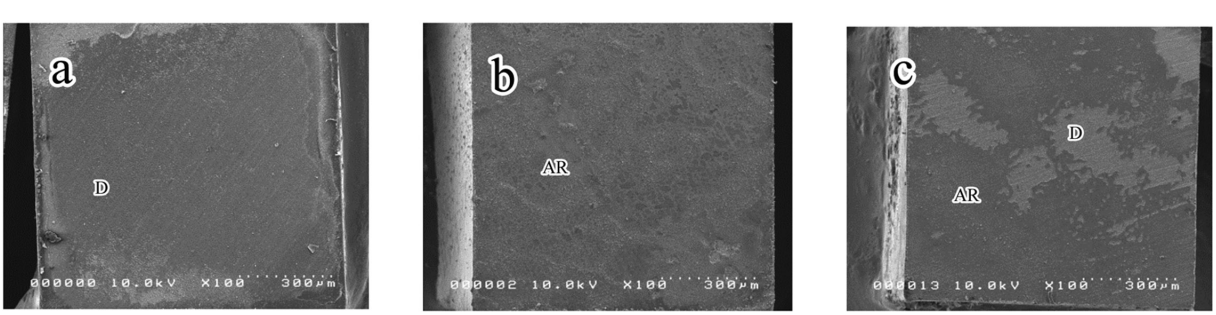

Fig. 2 Field-emission scanning electron micrographs of de-bonded samples show their fractured surfaces. (a) adhesive; (b) cohesive; (c) mixed. Abbreviations: adhesive resin (AR) ; dentin (D).

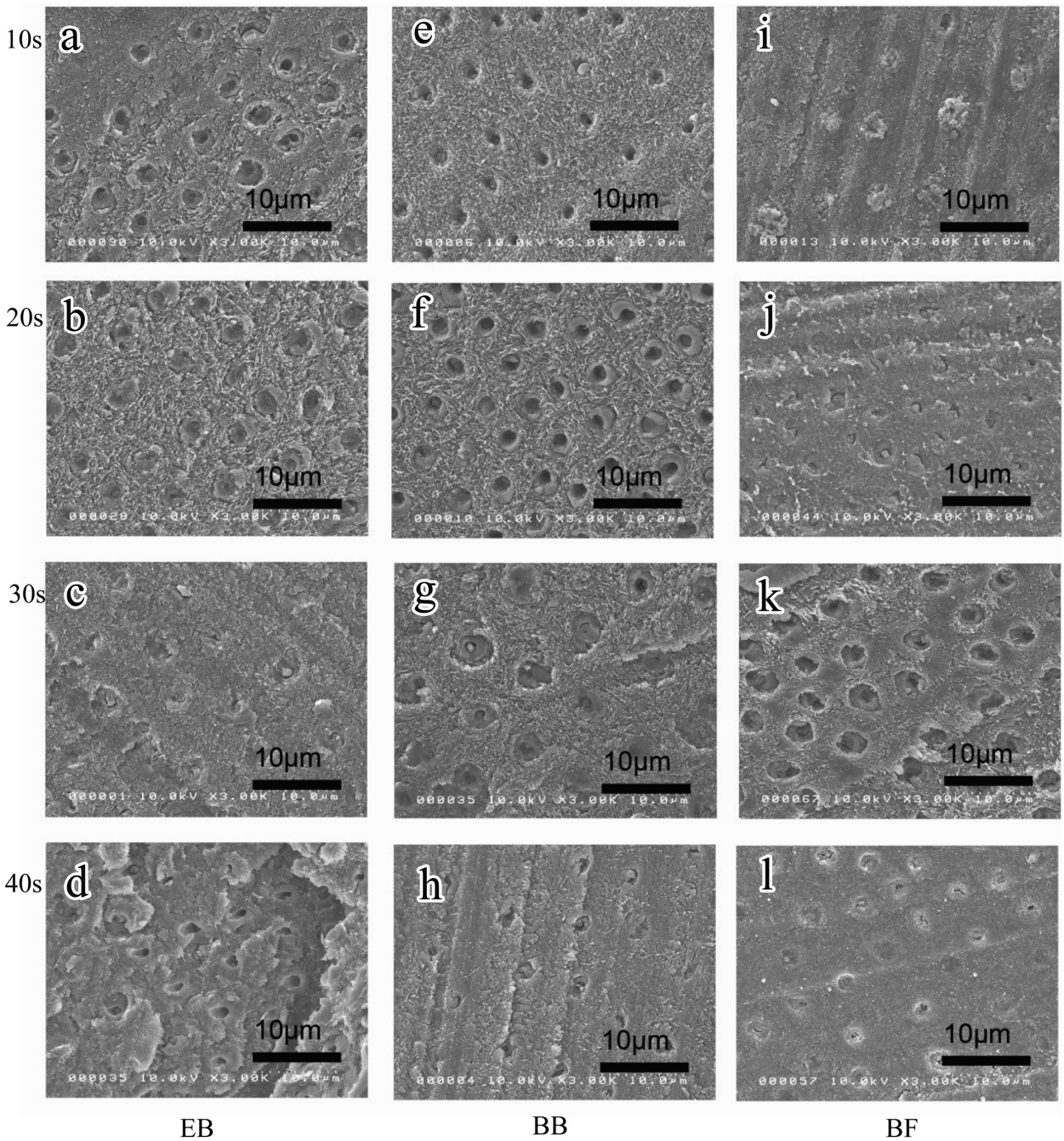

Fig. 3 SEM observations of the dentin surface from three systems

SEM observations of the dentin surface from three systems: (a) EB10s; (b) EB20s; (c) EB30s; (d) EB40s; (e) BB10s; (f) BB20s; (g) BB30s; (h) BB40s; (i) BF10s; (j) BF20s; (k) BF30s; (l) BF40s

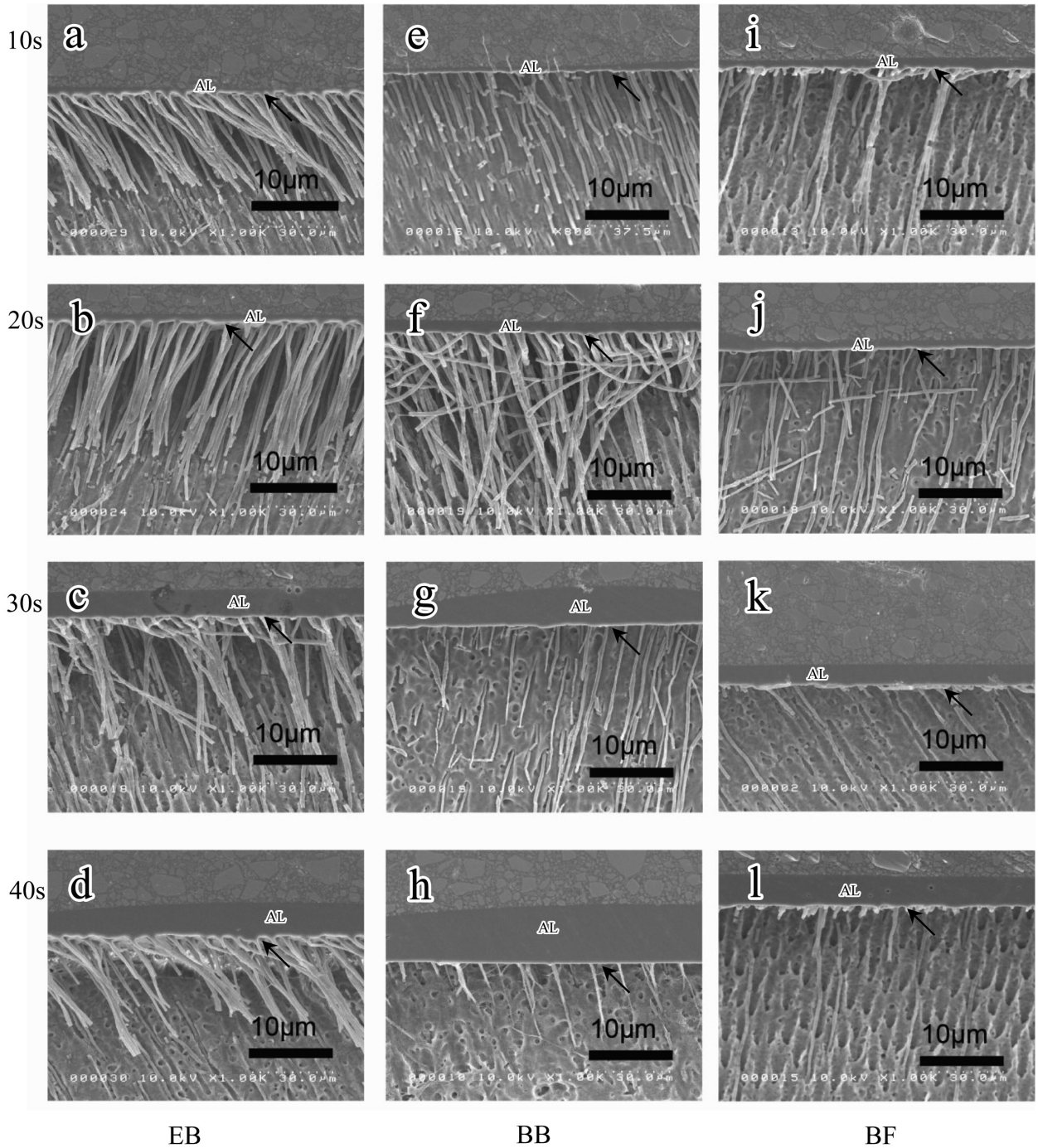

Fig. 4 The SEM observations for resin-dentin interface.
The SEM observations for resin-dentin interface: (a) EB10s; (b) EB20s; (c) EB30s; (d) EB40s; (e) BB10s; (f) BB20s; (g) BB30s; (h) BB40s; (i) BF10s; (j) BF20s; (k) BF30s; (l) BF40s. Abbreviations :Adhesive layer (AL); hybrid layers (arrowed).

DISCUSSION

By avoiding a separate etching step, the self-etch adhesive system greatly simplifies the procedure of operation by integrating the etching, priming and bonding step together. The basic principle is that their acidic functional components dissolve the smear layer and demineralize the tooth structure. However, the smear layer is not fully eliminated. The dissolved smear layer and adhesives monomers forms a hybrid layer and a special dentinal plug hybridized with resin. The hybrid layer and the dentinal plug become a part of resin tags to make bonding[13, 14].

The bonding strength of self-etch adhesive system

mainly comes from the formation of hybrid layer and resin tags. Weak acid etching leads to the demineralization of superficial dentin layer and the formation of a thinner hybrid layer[15, 16]. From the adhesive mechanism of self-etch adhesive system, there are many factors influence adhesives. Besides, the composition, the application number and time of self-etch adhesives may influence their bond strength. In order to avoid the interference of these factors in our study, we applied in a single direction and for only one coating.

The result of this study indicated that the application time of three adhesives had an effect on their bonding strength. When adhesives were applied for 40s, the strength reached to its lowest value compared with 10s, 20s, and 30s. It may be caused by the thicker bonding layer, which contained the droplets. The origin of these droplets is thought to be caused by an osmotic process precipitated by a gradient imbalance between water-rich dentin and water-poor oxygen-inhibition layer before and after light curing[17, 18]. Due to their inherent hydrophilicity, One-step self-etch adhesives are more prone to water uptake than more hydrophobic etch-and-rinse adhesives[19]. In these adhesives, water from the dentin diffuses through the hybrid layers and adhesive layers and form blisters after being cured, because they act as semi-permeable membranes[20]. This blister-rich zone jeopardizes copolymerization of monomers from the adhesive and the composite resin, resulting in a weaker interface and leading to low bond strength values.

According to the test results of three adhesives, the bond strength in 20s application groups were tended to show higher than that in 10s groups, but there was no statistically significant difference between them. Since all the three adhesives are acidic and their PH value is between 2 and 3, the extra application time would increase the concentration of acidic functional monomers and provide more solvent to transport these monomers into the matrix through the smear layer[11, 21-23]. Therefore, by adding acidic co-monomers on incompletely demineralized smear layers, further dis-solution and infiltration should be expected with the increase of adhesive layer thickness, consequently resulting in the enhancement of bond strength.

The bond strength declined when the adhesives were applied for 30s, probably because while diffusing through the deep channels within the bulk of the thick smear layer, the acidic monomers are rapidly neutralized, and fewer active monomers are left when they reach the underlying intact dentin. The long path through the smear layer probably prevented a sufficient concentration of active functional monomers to interact with the underlying dentin and produced an adequate hybrid layer. Therefore, the hybrid layer became weak and the bond strength decreased. Some studies show that the cured adhesive layer in all-in-one adhesives may act as semipermeable membranes that allow water diffusion from the bonded hydrated dentine to the intermixed zone between the adhesive and the uncured composite. Osmotic blistering of water droplets along the surface of the cured adhesive layer and emulsion polymerisation of immiscible resin components probably account for the compromised bond strength in single-step adhesives after the excessive application time[17].

By prolonging the application time, we discovered that the hybrid layers did not increase in thickness. Therefore, application time might change the quality of hybrid layer rather than its thickness. When adhesives were applied for 20s, resin tags had better continuity and regular shape mainly because the increase of permeability. Along with the increase of application time, the sufficient interaction between acidic monomers and dentinal surface led to the increase of the quality of hybrid layer which improve the permeability of adhesives. However, when adhesives were applied of 30s and 40s, the resin tags decreased in number, and many of them were incomplete, and the gaps emerged. It was primarily because along with the increase of application time, the ability of penetration of acidic monomers declined, the inadequate interaction between acid monomers and dentinal surface resulted in the decrease of the quality of the hybrid layer, and finally caused the mobility and penetration of resins decreased, and gaps emerged.

In comparison, the groups of EB obviously performed higher bond strength than others in each different application time, which is not shown in Table 2. Firstly, all the adhesives used the same solvents and had the similar acidity. The differences between them may rely on the composition and their ability of infiltration of superficial. Both EB and BF contains HEMA, which is the most essential component in adhesives. Due to the hydrophilicity, HEMA improves the wettability of resins and there by improves bond strength of adhesives. Therefore, EB had higher bond strength than BF. The reason of weaker bond strength in BF is perhaps the higher of concentration of HEMA brings out the liquid in dentinal tubules, resulting in the decreased sealing of

Table 2 Means of μTBS in MPa

adhesive	10s	20s	30s	40s
EB	44.43 ± 13.54^{ac}	50.12 ± 9.26^{a}	37.25 ± 6.66^{c}	22.51 ± 9.94^{d}
BB	30.21 ± 11.69^{MN}	37.32 ± 11.58^{M}	21.15 ± 8.42^{NP}	15.07 ± 8.58^{P}
BF	18.34 ± 7.13^{XY}	29.27 ± 8.31^{X}	15.12 ± 6.12^{Y}	12.87 ± 4.43^{Y}

Mean±standard deviation in MPa. The maximum value of each adhesive at 20s in different application time. The same letter are not statistically difference (p>0.05).

tubules and weaker bond strength. The bond strength of BF was even lower than that of BB, which did not include HEMA[24].

The above findings above show that the correct use of bonding materials is very important in dental restoration. When using self-etch adhesives, adequate increase of the application time properly can not only improve the bond strength, but also the sealing ability of dentinal tubules.

CONCLUSION

The null hypothesis that different application time has no effect on the bond strength of in all-in-one systems is false and rejected. Our result showed that the bond strength could increase when adhesives were applied for 20s.

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