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Influence of Etching Mode on Enamel Bond Durability of Universal Adhesive Systems

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Clinical Relevance

Total-etch mode has a positive effect on the enamel bond durability of universal adhesives, as was seen with the previous generation of single-step adhesives.

SUMMARY

The purpose of this study was to determine the enamel bond durability of three universal adhesives in different etching modes through fatigue testing. The three universal adhesives used were Scotchbond Universal, Prime&Bond Elect universal dental adhesive, and All-Bond Universal light-cured dental adhesive. A single-step self-etch adhesive, Clearfil S^3 Bond Plus was used as a control. The shear bond strength (SBS) and shear fatigue strength (SFS) to human enamel were evaluated in total-etch mode and self-etch mode. A stainless steel metal ring with an internal diameter of 2.4 mm was used to bond the resin composite to the flat-ground (4000-grit) tooth surfaces for determination of both SBS and SFS. For each enamel surface treatment, 15 specimens were prepared for SBS and 30 specimens for SFS. The staircase method for fatigue testing was then used to determine the SFS of the resin composite bonded to the enamel using 10-Hz frequencies for 50,000 cycles or until failure occurred. Scanning electron microscopy was used to observe representative debonded specimen surfaces and the resin-enamel interfaces. A two-way analysis of variance and the Tukey

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***post hoc* test were used for analysis of the SBS data, whereas a modified *t*-test with Bonferroni correction was used for the SFS data. All adhesives in total-etch mode showed significantly higher SBS and SFS values than those in self-etch mode. Although All-Bond Universal in self-etch mode showed a significantly lower SBS value than the other adhesives, there was no significant difference in SFS values among the adhesives in this mode. All adhesives showed higher SFS:SBS ratios in total-etch mode than in self-etch mode. With regard to the adhesive systems used in this study, universal adhesives showed higher enamel bond strengths in total-etch mode. Although the influence of different etching modes on the enamel-bonding performance of universal adhesives was found to be dependent on the adhesive material, total-etch mode effectively increased the enamel bond strength and durability, as measured by fatigue testing.**

INTRODUCTION

In the last decade, self-etch adhesive systems have become prevalent due to the reliability of bonding performance to the tooth structure. In addition, due to chemical bonding to hydroxyapatite and decreased demineralization of dentin, it has been claimed that the incidence of postoperative sensitivity is lower than for etch-and-rinse systems.[1,2] Generally, self-etch adhesive systems can be categorized as either two-step or single-step systems[3]; single-step self-etch adhesives are much easier to use, given that the priming and bonding procedures are combined into one step. This reduction in bonding procedures is beneficial for dental professionals because it allows for reduction in the technique sensitivity in clinical situations.

Recently, a new type of single-step self-etch adhesive categorized as "universal" or "multi-mode" has been introduced.[4-7] Universal adhesives can be used with multiple substrates, such as enamel, dentin, silica-based glass ceramics, zirconia ceramics, and metal alloys, without individual pretreatment.[8,9] This type of multifunctional adhesive is expected to help simplify the bonding procedure when various restorations are bonded to the tooth structure. Furthermore, some universal adhesives use a dual-cure system to allow for better polymerization at the interface and subsequent higher bond strengths when in contact with resin cements. The chemical reaction promoted by the activator may increase the bonding performance when adequate light irradiation is not possible.

For previous generations of single-step self-etch adhesives, some laboratory studies[10,11] have reported lower bonding performance than two-step self-etch systems and etch-and-rinse adhesive systems. In particular, enamel bond durability has been claimed to be a cause for concern due to the poorer mechanical properties of the adhesive layer and lower etching capability.[12] Therefore, to obtain a durable bond to the enamel, selective etching with phosphoric acid prior to application of the single-step self-etch adhesive has been recommended.[13-16] However, in practice, it is extremely difficult to precisely etch only the enamel surface, resulting in the strong possibility of inadvertently overetching exposed dentin. This may lead to decreased dentin bonding quality due to incomplete penetration of the resin monomers into demineralized dentin as well as induced postoperative sensitivity.[1,17] To overcome the weakness of previous generations of single-step self-etch adhesives, universal adhesives have been developed that allow for application of the adhesive with phosphoric acid pre-etching in the total-etch or selective-etch approaches.[4-7]

Although there are several laboratory and clinical studies regarding the bonding performance of universal adhesives,[18,19] only limited information is available on the bond durability of universal adhesives when used in different application modes. To understand the characteristics of universal adhesives, investigation of bonding performance from the perspective of fatigue stress, simulating the oral environment, is important because it clarifies the long-term bond durability. In addition, simulated oral environment testing can provide a rapid and standardized method to examine relative bond durability among materials and help predict expected clinical performance.[15,16]

The purpose of this laboratory investigation was to determine the enamel bond durability of universal adhesives in different application modes through fatigue testing. The null hypotheses to be tested were as follows: 1) universal adhesives would not differ from a single-step self-etch adhesive with respect to bond durability; 2) phosphoric acid pre-etching would not affect the enamel bond durability of universal adhesives.

METHODS AND MATERIALS

Materials

The materials used in this study are shown in Table 1. The following three universal adhesives were

Table 1: *Materials Used in This Study*

Code	Adhesive (Lot No.)	Main Components	Manufacturer
SU	Scotchbond Universal (41256)	MDP, HEMA, dimethacrylate resins, Vitrebond copolymer, filler, ethanol, water, initiators, silane	3M ESPE, St Paul, MN, USA
PE	Prime & Bond Elect (140710)	Dipentaerythritol pentaacrylate monophosphate, polymerizable dimethacrylate resin, polymerizable trimethacrylate resin, diketone, organic phosphine oxide, stabilizers, cetylamine hydrofluoride, acetone, water	Dentsply Caulk Milford, DE, USA
AU	All-Bond Universal (1300008503)	MDP, bis-GMA, HEMA, ethanol, water, initiators	Bisco Inc, Schaumburg, IL, USA
SP	Clearfil S^3 Bond Plus (4G0011)	MDP, bis-GMA, HEMA, ethanol, water, filler, CQ	Kuraray Noritake Dental Tokyo, Japan
	Pre-etching Agent		
	Ultra-Etch (G017)	35% phosphoric acid	Ultradent Products, Inc, South Jordan, UT, USA
	Resin composite		
	Z100 Restorative (N416713)	Bis-GMA, TEGDMA, silane treated ceramic, 2-benzotriazolyl-4-methyphenol catalysts, accelerators, CQ, pigments, others, zirconia/silica, 0.01-3.5 μm. Filler Load: 84.5% weight, 66% volume	3M ESPE, St Paul, MN, USA

Abbreviations: Bis-GMA, 2,2-bis[4-(2-hydroxy-3-methacryloyloxy propoxy) phenyl] propane; CQ, dl-camphorquinone; HEMA, 2-hydroxyethyl methacrylate; MDP, 10-methacryloyloxydecyl dihydrogen phosphate; TEGDMA, triethyleneglycol dimethacrylate.

used: Scotchbond Universal adhesive (SU; 3M ESPE, St Paul, MN, USA), Prime&Bond Elect universal dental adhesive (PE; Dentsply Caulk, Milford, DE, USA), and All-Bond Universal light-cured dental adhesive (AU; Bisco, Schaumburg, IL, USA). A conventional single-step self-etch adhesive, Clearfil S^3 Bond Plus (SP; Kuraray Noritake Dental, Tokyo, Japan), was used as a control. Ultra-Etch (Ultradent, South Jordan, UT, USA) was used as a 35% phosphoric acid etching agent. A visible light-cured resin composite (Z100 Restorative, 3M ESPE) was used as a restorative material for bonding to enamel.

Specimen Preparation

Deidentified, extracted, caries-free human molars were selected for use in this study under a protocol reviewed and approved by the Ethics Committee for Human Studies of the Nihon University School of Dentistry (No. 2015-06). The enamel bonding sites were prepared by mesiodistally sectioning the teeth and removing approximately two-thirds of the apical root structure. The buccal and lingual tooth sections were mounted with a resin material (Triad DuaLine, Dentsply Trubyte, York, PA, USA) in 25-mm-diameter aluminum rings. The enamel-bonding surfaces were ground flat to 4,000-grit using a water coolant and a series of carbide polishing papers (Struers Inc, Cleveland, OH, USA). Metal rings machined from SAE 304 stainless steel with an internal diameter of 2.4 mm, an external diameter of 4.8 mm, and a length of 2.6 mm were used to condense the resin composite on enamel surfaces for shear bond strength (SBS) and shear fatigue strength (SFS) tests. The bonding procedure resulted in a resin composite cylinder inside the ring that was approximately 2.36 mm in diameter and 2.5 mm in height. The ring was left in place for the tests.

SBS Tests

For each test group, 15 specimens were used to determine the SBS to enamel in total-etch mode (phosphoric acid was applied for 15 seconds prior to application of the adhesive) or self-etch mode (without phosphoric acid etching). The adhesive agents were used in accordance with the manufacturers' instructions, as shown in Table 2. Following treatment of the flat-ground enamel surface with the adhesive agent, the stainless steel metal ring was placed over the enamel surface and secured by clamping with a custom fixture. The light-cured resin composite paste was condensed in the ring and light-irradiated for 40 seconds with a curing unit (Spectrum 800, Dentsply Caulk, Milford, DE, USA) set at a light irradiance average of 600 mW/cm^2. The bonded specimens were stored for 24 hours in 37°C distilled water before testing. The specimens were loaded to failure at a rate of 1.0 mm/min using a universal testing machine (Electron E 1000, Instron, Norwood, MA, USA). A metal rod with a chisel-shaped end was used to apply the load to the metal

Table 2: *Application Protocol for Pre-etching and Self-etching Adhesives*

Method	**Pre-etching Protocol**
Total-etch	Enamel surface was phosphoric acid etched for 15 s, then rinsed with water for 15 s (three-way dental syringe) and air-dried.
Self-etch	Phosphoric acid pre-etching was not performed.
Adhesive	**Adhesive Application Protocol**
SU	Adhesive applied to air-dried enamel surface with rubbing for 20 s. Medium air pressure applied to surface for 5 s. Light-irradiated for 10 s.
PE	Adhesive applied to enamel surface with rubbing for 20 s. Gentle stream of air applied over the liquid for at least 5 s. Light-irradiated for 10 s.
AU	Adhesive applied to enamel surface with rubbing action for 10-15 s/coat. No light cure between coats. Gentle stream of air applied over the liquid for at least 10 s. Light-irradiated for 10 s.
SP	Adhesive applied to air-dried enamel surface for 10 s. Medium air pressure applied to surface for 5 s. Light-irradiated for 10 s.

Abbreviations: AU, All-Bond Universal light-cured dental adhesive; PE, Prime&Bond Elect universal dental adhesive; SP, Clearfil S^3 Bond Plus; SU, Scotchbond Universal.

ring immediately adjacent to the flat-ground enamel surface. The SBS values (MPa) were calculated from the peak load at failure divided by the bonded surface area. After testing, the bonding sites of the enamel surface and resin composite cylinders were observed using an optical microscope (MZ16; Leica Microsystems, Heerbrugg, Switzerland) at 20× magnification to determine the bond failure mode. On the basis of the percentage of substrate area (adhesive, resin composite, and enamel) observed on the debonded cylinders and tooth-bonding sites, the types of bond failure were recorded as 1) adhesive failure, 2) cohesive failure in composite, 3) cohesive failure in enamel, or 4) mixed failure (ie, partially adhesive and partially cohesive).

SFS Tests

The staircase method of fatigue testing reported by Draughn[20] was used for SFS testing, as previously reported.[15,16] Test specimens were prepared as described above for SBS testing. The lower load limit was set near zero (0.4 N), and the initial maximum load applied was 50%-60% of the SBS value determined for each of the adhesive systems tested. The load was applied at a frequency rate of 10 Hz with an ElectroPuls E1000 machine (Instron, Norwood, MA, USA) using a sine wave for 50,000 cycles or until failure occurred. Our preliminary investigations showed that the frequency had no significant effect on the results, and 10 Hz was selected for reasons of practicality.

The load was incrementally increased or decreased (depending on survival or failure) by approximately 10% of the initial load. For each test condition, 30 specimens were used to determine the SFS. Adapting the calculation described by Draughn,[20] the test stress that produces 50% failure is termed the fatigue strength. After testing, the specimens were examined to determine the type of bond failure in the same manner as for SBS, as described in the previous section.

Scanning Electron Microscopy Observations

Restorative-enamel interfaces and representative fracture sites after SFS testing were observed by field-emission scanning electron microscopy (SEM; ERA-8800FE, Elionix, Tokyo, Japan). For ultrastructural observations of the restorative/enamel interface, bonded specimens were embedded in epoxy resin and longitudinally sectioned with a precision low-speed saw (Isomet 111280, Buehler, Lake Bluff, IL, USA). The sectioned surfaces were polished to a high gloss with abrasive discs (Fuji Star Type DDC, Sankyo Rikagaku Co, Saitama, Japan) followed by diamond pastes to a 0.25-μm particle size (DP-Paste, Struers, Ballerup, Denmark). Fracture sites were prepared directly for evaporation coating. SEM specimens of restorative-enamel interfaces were dehydrated in ascending grades of tert-butyl alcohol (50% for 20 minutes, 75% for 20 minutes, 95% for 20 minutes, and 100% for 2 hours) and transferred from the final 100% bath to a critical-point dryer (Model ID-3, Elionix) for 30 minutes. Resin-enamel interface specimens were then subjected to argon-ion beam etching (EIS-200ER, Elionix) for 40 seconds with the ion beam (accelerating voltage 1.0 kV, ion current density 0.4 mA/cm^2) directed perpendicular to the polished surfaces. Finally, all SEM specimens were coated with a thin film of gold in a Quick Coater vacuum evaporator (Type SC-701, Sanyu Denchi Inc, Tokyo, Japan). Observations were carried out using an operating voltage of 10 kV.

Table 3: *Influence of Etching Mode on SBS in MPa (SD)*[a]

Adhesive	Total-etch Mode	Self-etch Mode
SU	46.4 (5.4)^{aA}	27.7 (3.8)^{bB}
PE	42.6 (5.2)^{aA}	28.8 (5.3)^{bB}
AU	42.1 (4.9)^{aA}	24.1 (2.4)^{cB}
SP	43.5 (5.5)^{aA}	27.5 (2.3)^{bB}

Abbreviations: AU, All-Bond Universal light-cured dental adhesive; PE, Prime&Bond Elect universal dental adhesive; SBS, shear bond strength; SFS, shear fatigue strength; SP, Clearfil S^3 Bond Plus; SU, Scotchbond Universal.
[a] Same lowercase letter in vertical columns indicates no difference at 5% significance level. Same capital letter in horizontal rows indicates no difference at 5% significance level.

Statistical Analysis

A two-way analysis of variance (ANOVA) followed by a Tukey honestly significant difference test at an α level of 0.05 were used for analysis of the SBS data. The statistical analysis for SBS was performed using the Sigma Plot software system (version 11.0; SPSS Inc, Chicago, IL, USA). A modified *t*-test with Bonferroni correction was used for the SFS data with a custom program implemented in a spreadsheet (Excel, Microsoft Inc, Redmond, WA, USA).

RESULTS

Shear Bond Strength

The SBS results in different etching modes are shown in Table 3. The two-way ANOVA revealed that the etching mode (total-etch vs self-etch) significantly influenced the SBS values ($p<0.001$), whereas the adhesive system did not ($p=0.390$). However, the interaction between these factors was significant ($p=0.003$).

The mean SBS in total-etch mode ranged from 42.6 ± 5.2 to 46.4 ± 5.4 MPa, whereas the corresponding values for the specimens in self-etch mode ranged from 24.1 ± 2.4 to 28.8 ± 5.3 MPa (Table 3). All adhesives showed significantly higher SBS values in total-etch mode than in self-etch mode. Although there was no significant difference among the tested adhesives in total-etch mode, AU showed a significantly lower SBS value ($p<0.05$) than the other adhesives in self-etch mode.

Shear Fatigue Strength

The SFS results in different etching modes are shown in Table 4. The mean SFS in total-etch mode ranged from 21.0 ± 4.8 to 22.4 ± 4.0 MPa, whereas the corresponding values for the specimens in self-etch mode ranged from 10.0 ± 2.2 to 12.8 ± 1.6 MPa. There was no significant difference among the tested adhesives ($p>0.05$) in both total-etch and self-etch modes. The ratios of SFS:SBS are shown in Table 4. For all adhesives, the total-etch groups demonstrated higher ratios than the self-etch groups.

Failure Mode Analysis of Debonded Specimens

The frequency of different failure modes, comparing the modes seen in SBS testing with those seen in SFS testing, are shown in Table 5. For all adhesives, the predominant mode of failure for specimens in self-etch mode was adhesive failure regardless of the testing methods. However, mixed failure and cohesive failure in enamel increased in total-etch mode for all adhesives in both SBS and SFS.

SEM Observations

Representative SEM images of debonded specimens after SFS testing are shown in Figures 1-4. For the universal adhesives, no clear morphological differences were observed between the self-etch mode and total-etch mode at lower magnifications (Figures 1a,c, 2a,c, and 3a,c). However, at higher magnification, cracks, cleavages, and cohesive failures in enamel could be seen more clearly in the total-etch group (Figures 1d, 2d, and 3d). For SP in self-etch mode, detachment of the boundary at the interface between the adhesive and enamel surface was observed at lower magnification (Figure 4a). Conversely, in total-etch mode, the de-

Table 4: *Influence of Etching Mode on SFS (MPa) and Ratio of SFS/SBS*[a]

Adhesive	Total-etch Mode	Self-etch Mode	Ratio SFS:SBS	
			Total-etch Mode	Self-etch Mode
SU	22.3 (4.6)^{aA}	12.8 (1.6)^{bB}	0.480	0.462
PE	21.0 (4.8)^{aA}	10.7 (1.2)^{bB}	0.493	0.372
AU	21.6 (2.2)^{aA}	10.0 (2.2)^{bB}	0.513	0.415
SP	22.4 (4.0)^{aA}	12.3 (3.5)^{bB}	0.515	0.447

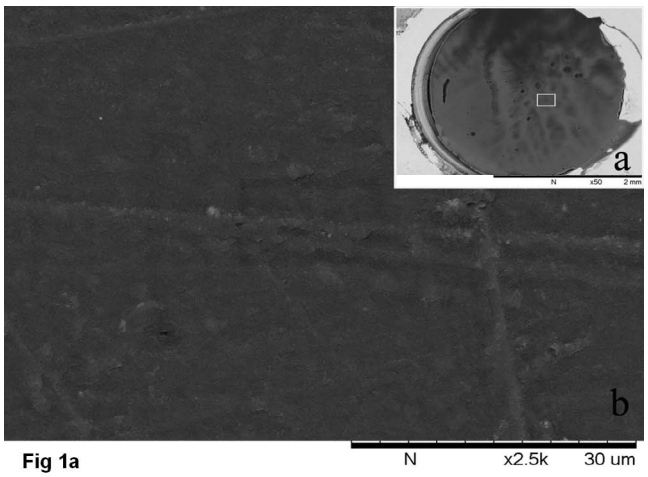
Abbreviations: AU, All-Bond Universal light-cured dental adhesive; PE, Prime&Bond Elect universal dental adhesive; SBS, shear bond strength; SFS, shear fatigue strength; SP, Clearfil S^3 Bond Plus; SU, Scotchbond Universal.
[a] Same lowercase letter in vertical columns indicates no difference at 5% significance level. Same capital letter in horizontal rows indicates no difference at 5% significance level.

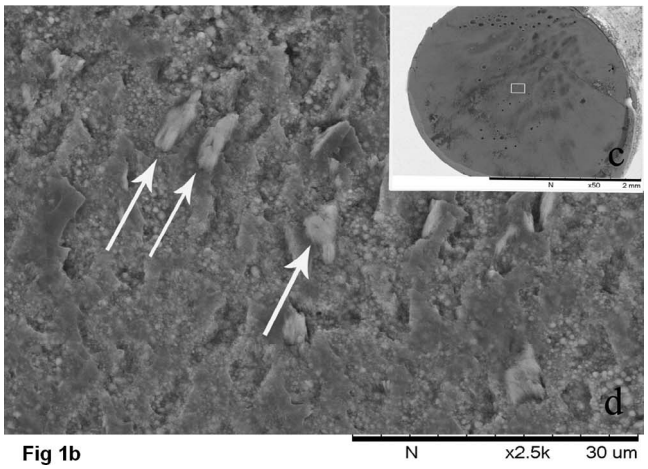
Table 5: *Failure Mode Analysis of Debonded Specimens*[a]

Adhesive	SBS		SFS	
	Total-etch	Self-etch	Total-etch	Self-etch
SU	[60/0/20/20]	[93/0/0/7]	[55/9/27/9]	[100/0/0/0]
PE	[80/0/0/20]	[100/0/0/0]	[77/0/23/0]	[100/0/0/0]
AU	[79/7/7/7]	[100/0/0/0]	[58/0/21/21]	[100/0/0/0]
SP	[67/0/26/7]	[100/0/0/0]	[77/0/23/0]	[100/0/0/0]

Abbreviations: AU, All-Bond Universal light-cured dental adhesive; PE, Prime&Bond Elect universal dental adhesive; SBS, shear bond strength; SFS, shear fatigue strength; SP, Clearfil S^3 *Bond Plus; SU, Scotchbond Universal.*
[a] *Failure mode: [adhesive failure/cohesive failure in resin composite/cohesive failure in enamel/mixed failure] percentage of each failure mode.*

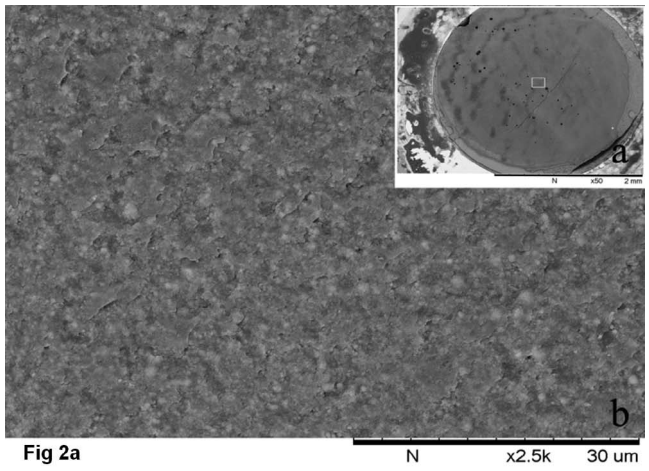
tachment area at the interface between the adhesive and resin composite increased, compared with self-etch mode (Figure 4c). In addition, cohesive failures in enamel and beach marks were more clearly visible (Figure 4d) than in self-etch mode (Figure 4b).





Figures 1-4. *Representative scanning electron microscope images of the fractured resin surface in enamel bonding after SFS.*
Figure 1. *(a): Scotchbond Universal in total-etch mode. (b): Scotchbond Universal in self-etch mode. a,c: 40×; b,d: 1000×.*

Representative SEM images of the resin-enamel interface are shown in Figures 5-8. Although clear differences were observed between samples of the two different treatment modes, the restorative-enamel interface showed excellent adaptation to the enamel surface regardless of the etching mode. However, the resin tags in the enamel surfaces were longer for specimens in total-etch mode than in self-etch mode. The thickness of the adhesive



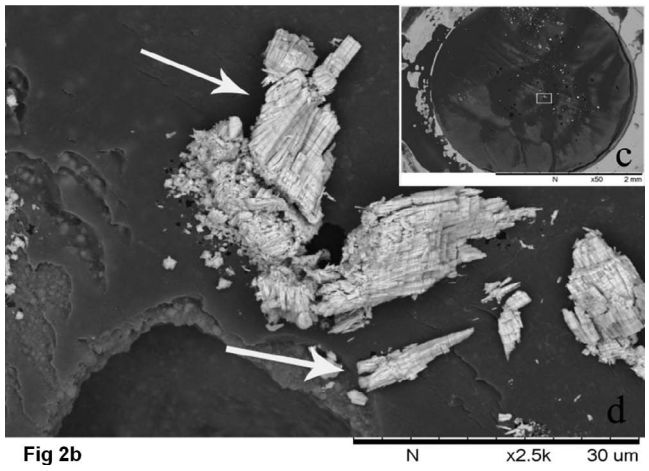


Figure 2. *(a): Prime&Bond elect in total-etch mode. (b): Prime& Bond elect in self-etch mode. a,c: 40×; b,d: 1000×.*

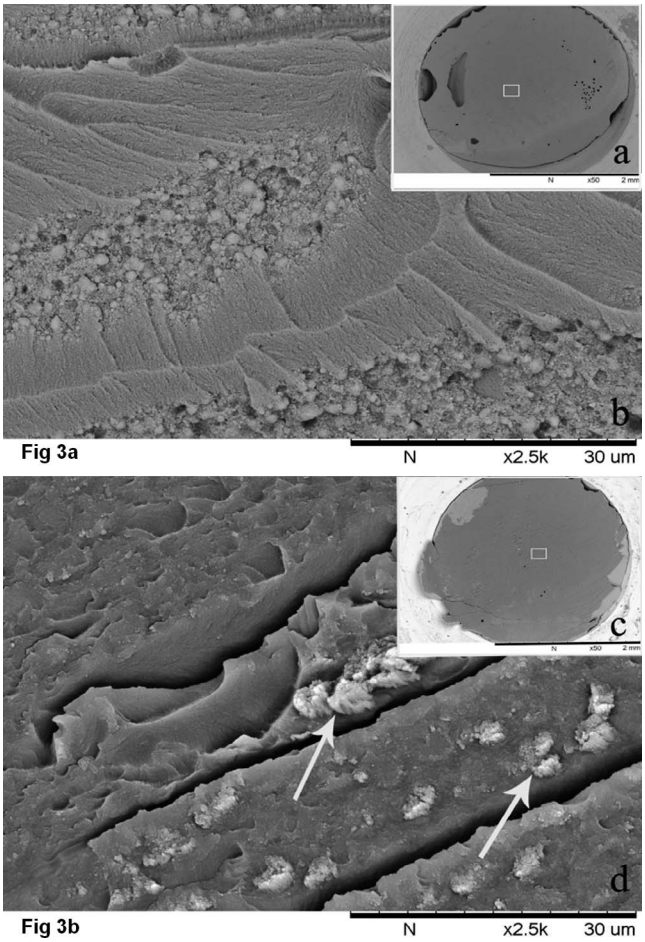


Figure 3. *(a): Clearfil S^3 Bond Plus in total-etch mode. (b): Clearfil S^3 Bond Plus in self-etch mode. a,c: 40×; b,d: 1000×. Arrows indicate cohesive failure in enamel.*

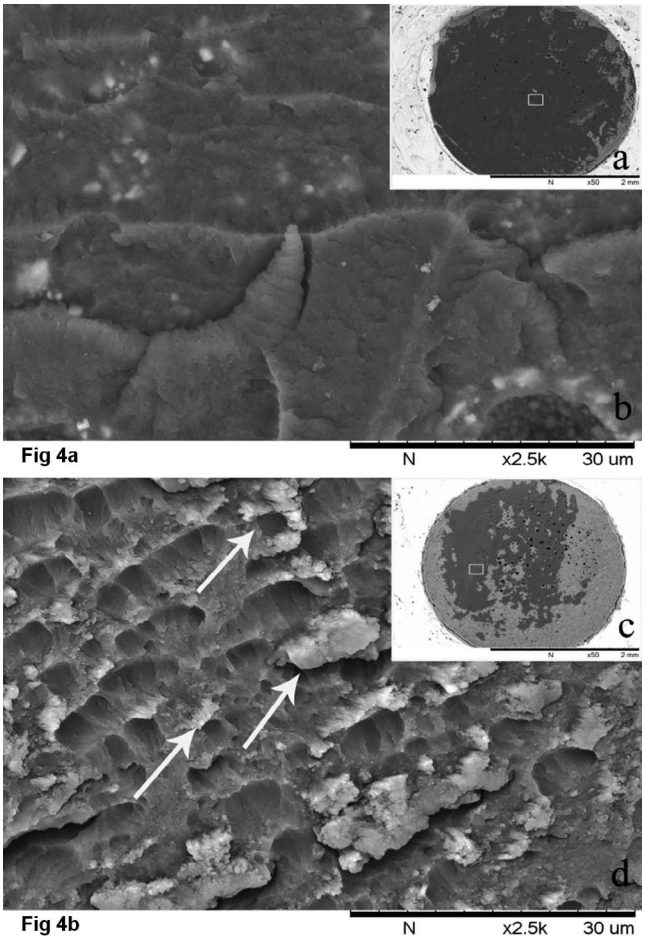


Figure 4. *(a): All-Bond Universal in total-etch mode. (b): All-Bond Universal in self-etch mode. a,c: 40×; b,d: 1000×.*

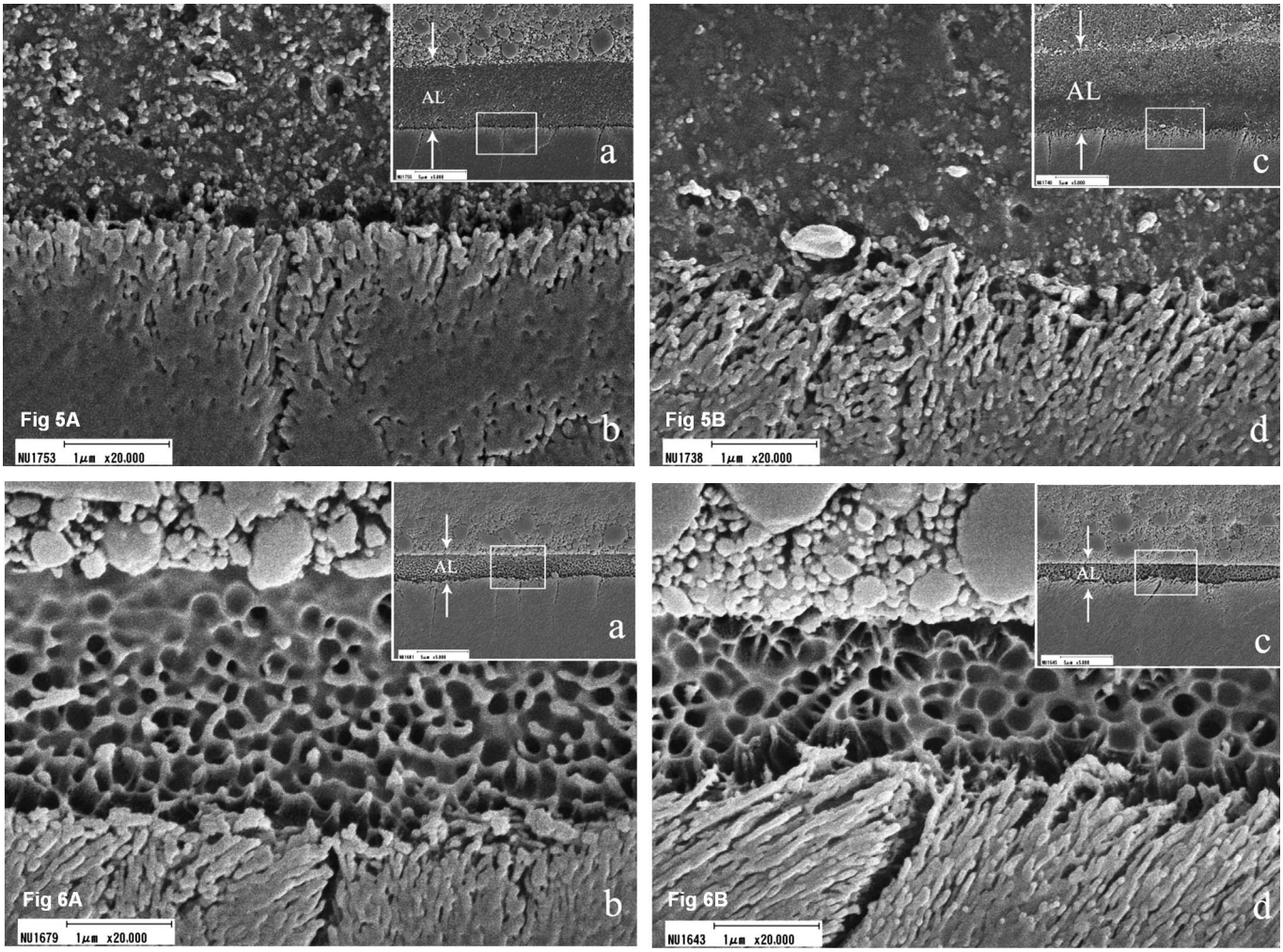
layer was 2-3 μm for PE (Figure 6) and 4-5μm for AU (Figure 7). The adhesive layers of SU (Figure 5) and SP (Figure 8) had similar thicknesses of 7-10 μm.

The nano-fillers in SU and SP (Figures 5 and 8) were clearly observed, although they were more prominent in SP. EL and AU did not contain nano-fillers (Figures 6 and 7) but had distinct adhesive layers. The adhesive layer of AU was largely homogeneous, with obvious inclusion of enamel fragments (arrows in Figure 7). Conversely, PE possessed a clear honeycomb network without enamel fragments (Figure 6).

DISCUSSION

Manufacturers of universal adhesives have claimed that there is no compromise in bonding performance when either total-etch or self-etch modes are used.[21] Multi-mode use of universal adhesives may address the disadvantages of previous generation single-step self-etch adhesives with lower enamel bond durability when used in self-etch mode. If true, this would allow the use of multiple modes in the same restoration. This, in turn, would allow clinicians to adapt appropriately to heterogeneous substrates, which would improve bond durability and reduce postoperative sensitivity. Similarly, when a single mode must be used for the whole of a small restoration, any heterogeneity of the substrate should not have a negative effect. This should have clinical benefits.

However, there is concern that the technology of universal adhesives may not offer a genuine advantage when compared with previous generations of single-step self-etch adhesives.[7,21] In addition, to adapt to multiple substrates without individual pretreatment, the composition of universal adhesives is more complicated than that of the previous generation of self-etch adhesives. However, there is minimal information regarding the influence of



Figures 5-8. *Representative scanning electron microscope images of the resin-enamel interface. The visible material is indicated by the abbreviation AL: adhesive layer.*
Figure 5. *(a): Scotchbond Universal in total-etch mode. (b): Scotchbond Universal in self-etch mode. a,c: 5000×; b,d: 20,000×.*
Figure 6. *(a): Prime&Bond elect in total-etch mode. (b): Prime&Bond elect in self-etch mode. a,c: 5000×; b,d: 20,000×.*

different application modes on the durability of the bond to tooth substrates. Therefore, to clarify the bonding performance of universal adhesives, enamel bond durability was evaluated through fatigue testing.

In self-etch mode, AU showed a significantly lower SBS value than the other adhesives. However, in total-etch mode, there were no significant differences among the adhesives in SBS. The reason that AU demonstrated a lower SBS value in self-etch mode is thought to be related to it having a lower etching capability than other universal adhesives. Chen and others[21] reported that PE and SU are classified as mild self-etch adhesives, whereas AU is classified as an ultramild self-etch adhesive on the basis of pH and transmission electron microscope observations of the adhesives. Therefore, the lower acidity of AU might not allow for strong micromechanical retention, leading to a lower SBS value.

There were no significant differences among all adhesives in SFS, although values in self-etch mode were significantly lower than in total-etch mode. In particular, none of the universal adhesives differed from the conventional single-step self-etch adhesive SP. Therefore, the null hypothesis that universal adhesives would not differ from conventional single-step self-etch adhesives with respect to bond durability was not rejected. However, the other null hypothesis (that phosphoric acid pre-etching would not affect the enamel bond durability of universal adhesives) was rejected.

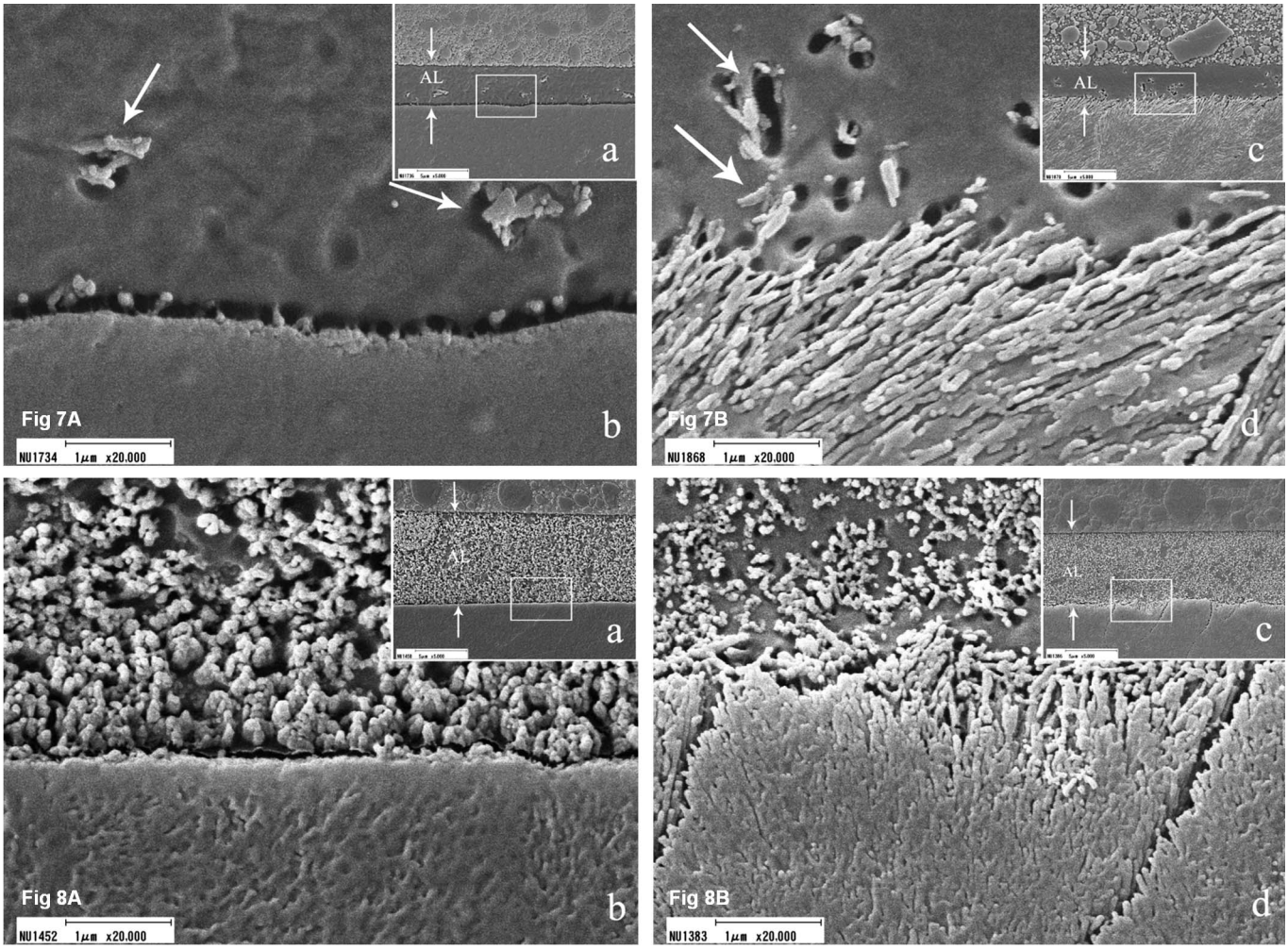


Figure 7. *(a): All-Bond Universal in total-etch mode. (b): All-Bond Universal in self-etch mode. a,c: 5000×; b,d: 20,000×. Arrows indicate enamel fragments in adhesive layers.*
Figure 8. *(a): Clearfil S^3 Bond Plus in total-etch mode. (b): Clearfil S^3 Bond Plus in self-etch mode. a,c: 5000×; b,d: 20,000×.*

The durability of bonded interfaces is threatened by many factors, such as biofilm attack, hydrolytic degradation of the adhesive, and fatigue of the adhesives.[23-25] In addition, dentin bonded interfaces are attacked through enzymatic degradation by matrix metalloproteinases.[26,27] Conversely, unlike dentin, enamel is composed of homogeneous components with minimal organic content. Therefore, the process of enamel bond degradation is simpler than that of dentin bonds due to the absence of collagen fibrils. Hence, deterioration of the adhesive layer in enamel bonds may directly lead to failure of the bond itself, indicating that the composition and properties of the adhesive layer are the primary factors in enamel bond durability.

Fatigue can be defined as the degradation or failure of mechanical properties after repeated subcritical stress at a level below the ultimate fracture strength of the material or interface.[22] Fatigue testing carried out in this study provided not only information related to the endurance characteristics of the bonding systems but also information regarding the ability of the resin-enamel interface zone to resist stress loading. When considering long-term bond durability, the degradation mechanism of the resin-tooth interface is extremely important.

Although there was no statistically significant difference in SFS among the adhesives, AU and PE tended to show lower values than did SU and SP. Although no clinical conclusions can be drawn from this difference because it is not significant, the compositions of the adhesives suggest that these differences may be valid and worthy of further investigation. The adhesive layers of SU and SP contain nano-fillers and are approximately two to three times thicker than those of AU and PE. This

thicker adhesive layer may behave as a shock absorber and the presence of nano-filler may inhibit crack propagation. In addition, the thicker adhesive layer may decrease the relative importance of the oxygen inhibition layer, which is thought to be a vulnerable site.[28] It can thus be proposed that different characteristics of the adhesive layers influenced the enamel bond durability in self-etch mode.

Erickson and others[29] reported that self-etch adhesives produce an etching pattern primarily involving the ends of enamel prisms and fine pitting of the enamel surface, with minimal effect on the interprismatic regions. Bond strength to phosphoric acid–etched enamel is mainly attributable to the penetration of adhesives into the enamel crystals and rods. From the SEM observations of enamel-resin interfaces in self-etch mode, gaps between enamel crystals and rods were not found in any adhesive. However, in total-etch mode, spicular etching patterns and penetration of resin tags were clearly observed for all adhesives. In addition, from the SFS results, total-etch mode significantly increased the enamel bond durability of universal adhesives compared with self-etch mode. Therefore, creating micromechanical retention on the enamel surface through phosphoric acid pre-etching may contribute to better resistance of long-term biomechanical loads when using universal adhesives.

Regarding dentin bonding, we evaluated the influence of different etching modes on the dentin bond durability of three universal adhesives using the same fatigue testing method as the present study.[30] In our study, the SBS and SFS of dentin bonds of a conventional single-step self-etch adhesive showed significantly lower values in total-etch mode when compared with those in self-etch mode. In contrast, the SBS and SFS of the universal adhesives showed equivalent bonding quality to dentin regardless of etching mode, although the dentin bond quality of the universal adhesives did vary among adhesives. We concluded that using universal adhesives in total-etch mode does not have a negative impact on dentin bond quality. However, postoperative sensitivity arises from a different mechanism from bond strength, and increased etching of dentin in the total-etch mode may increase it. This is an important topic for further work.

The clinical implication of this study is that universal adhesives should be used with phosphoric acid pre-etching of enamel, as with the previous generation of single-step self-etch adhesives. In clinical situations, the adherent surface of mineralized tooth tissue is dependent on the cavity configuration. Given that there is no compromise in bonding performance when either total-etch or self-etch mode is used with dentin, total-etch or selective-etch mode should be chosen to achieve a reliable bond to the enamel substrate. In particular, if the cavity is too small to precisely manipulate the phosphoric acid agent for selective etching, the total-etch approach with universal adhesives is most likely appropriate.

CONCLUSIONS

Within the limitations of this *in vitro* study, although AU demonstrated a significantly lower SBS value than the other tested adhesives in self-etch mode, there were no significant differences among the adhesives in both SBS and SFS in total-etch mode and in SFS in self-etch mode. SEM observations of resin-enamel interfaces revealed that the resin tags in enamel surfaces were longer for the samples in total-etch mode than those in self-etch mode. In addition, the thickness and characteristics of the adhesive layers were dependent on the adhesive.

All tested adhesives showed significantly higher values in total-etch mode than in self-etch mode. Therefore, for universal adhesives, total-etch mode has a positive effect on the enamel bond durability, as with the previous generation of single-step adhesives.

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Regulatory Statement

This study was conducted in accordance with all the provisions of the local human subjects oversight committee guidelines and policies of Nihon University School of Dentistry. The approval code for the study is 2015-06.

Conflict of Interest

The authors of this manuscript certify that they have no proprietary, financial, or other personal interest of any nature or kind in any product, service, and/or company that is presented in this article.

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