Influence of different etching modes
on dentin bonding effectiveness of universal adhesives

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Contents

This thesis was structured on the basis of the published article listed below, with additional data using Clearfil Universal Bond Quick (CU, Kuraray Noritake Dental, Tokyo, Japan).

Yamauchi K, Tsujimoto A, Jurado CA, Shimatani Y, Nagura Y, Takamizawa T, Barkmeier WW, Latta MA, Miyazaki M (2019) Etch-and-rinse vs self-etch mode for dentin bonding effectiveness of universal adhesives. J Oral Sci 61, 549-553.

Summary

Recently, universal adhesives, which can be used in any of etch-and-rinse, self-etch or selective-etch modes, have been developed. Although some reports have suggested that the bond durability of universal adhesives is inferior to that of two-step self-etch adhesives, clinical usage of universal adhesives is rapidly increasing due to their versatility. It is therefore important to thoroughly characterize the properties and behavior of these adhesives. Although it has been shown that universal adhesives have similar bonding performance regardless of the bonding strategy employed, more detailed analyses are still required. The purpose of this laboratory study was to assess the dentin bond fatigue resistance and interfacial characteristics of universal adhesives in etch-and-rinse and self-etch modes.

The five universal adhesives used were Adhese Universal (Ivoclar Vivadent, Schaan, Liechtenstein), All-Bond Universal (Bisco, Schaumburg, IL, USA), Clearfil Universal Bond Quick (Kuraray Noritake Dental, Tokyo, Japan), G-Premio Bond (GC, Tokyo, Japan) and Scotchbond Universal Adhesive (3M Oral Care, St. Paul, MN, USA). The dentin surfaces were treated with the universal adhesives in either etch-and-rinse or self-etch mode. A custom fixture was used to position and hold stainless steel rings over the bonding sites as the resin composite was placed into the rings. The resin composite was light-cured for 40 s. The bonded specimens were stored in 37ºC distilled water for 24 h before testing. Fifteen specimens per test group were loaded to failure using an all-electric dynamic instrument with a chisel-shaped metal load at a crosshead speed of 1.0 mm/min, and initial bond strength was obtained. The fatigue load was applied to the metal rings using a sine wave at a frequency of 20 Hz for 50,000 cycles or until failure occurred. The lower load limit was set at 0.4 N, and subsequent loading was adjusted upward or downward approximately 10% from the previous load depending on specimen survival

or failure. The surface free energy characteristics of adhesive treated dentin were determined by measuring the contact angle formed with the surface by the three test liquids 1-bromonaphthalene, diiodomethane, and distilled water. SEM observations of resin-dentin interfaces of the adhesives for etch-and-rinse and self-etch modes were also conducted.

The results of this study showed that etching mode did not affect the bond fatigue resistance of universal adhesives to dentin. A similar bonding study to enamel reported that the bond fatigue resistance of universal adhesives was significantly higher in etch-and-rinse than in self-etch mode. Therefore, the use of etch-and-rinse or selective etching modes with universal adhesives appears more effective. Results for the surface free energy characteristics of the baseline showed that γs, γs^d, γs^p, and γs^h of dentin in etch-and-rinse mode were significantly lower than those in self-etch mode. This means that phosphoric acid etched dentin has lower wettability. In the SEM observation of adhesive interfaces in etch-and-rinse mode, deeper penetration of adhesives into the dentinal tubules was observed due to the removal of the smear layer and opening of the dentinal tubules by phosphoric acid etching. These deeper resin tags may contribute to the higher resistance of adhesive interfaces to cyclic stress.

Taken together, these results suggest that the dentin bonding mechanisms in universal adhesives may vary substantially between adhesives, and between etching mode for the same adhesive. A better understanding of these mechanisms may allow significant improvements to be made to the dentin bonding performance of universal adhesives, and thus they are an important topic for further research.

Introduction

Recently, universal adhesives have become popular in dentistry because they can be used in any of etch-and-rinse, self-etch, or selective-etch modes (1). With the expiration of the patent for 10-methacryloyloxydecyl dihydrogen phosphate (10-MDP), held by Kuraray Noritake Dental (Tokyo, Japan), in 2003, manufacturers began exploring the usage of 10-MDP combination with other components in novel adhesive formulations. Scotchbond Universal Adhesive (3M Oral Care, St. Paul, MN, USA) was the first universal adhesive introduced commercially in Japan, in November of 2012. Later, universal adhesives that could be used with resin luting cements on various substrates (2), with reduced application times (3), or with various surface moisture conditions on enamel and dentin substrates (4) were brought onto the market. On the other hand, one report suggested that the bond durability of universal adhesives is inferior to that of two-step self-etch adhesives (5). Nevertheless, the flexibility of universal adhesives has ensured increasing popularity in the clinics (6), creating a need to further investigate better ways to utilize universal adhesives.

Over the past decade, a method designed to assess bond fatigue resistance in adhesives has been developed in a collaborative effort between the Nihon University School of Dentistry (Tokyo, Japan) and Creighton University School of Dentistry (Omaha, NE, USA). Development started with Erickson et al. (7) and was refined by researchers from NU and CU (8-10). The appropriate frequency (9), number of cycles (10), and analysis (11) are now established. However, at the present time, research using this bond fatigue test to compare the bonding performance of universal adhesives in etch-and-rinse and self-etch mode is limited.

Yoshida et al. (12) reported that the chemical interaction of 10-MDP with dentin is essential in obtaining durable bonds. Previously, Inoue et al. (13) showed that, drawing from an interfacial science perspective, chemical bond interactions between 10-MDP and dentin could be explained through changes in dentin surface characteristics. Although it has been shown in the past that universal adhesives have similar bonding performance regardless of the bonding strategy employed (1), it remains possible that the chemical bonding interactions of universal adhesive with ground and etched dentin are different. In addition, further assessment of the changes of the energetic characteristics of a dentin surface treated with universal adhesives in the two different modes, when added to a bond fatigue resistance analysis, may provide an explanation for the discrepancy between bond strength and chemical bonding effectiveness.

The purpose of this laboratory study was to assess the dentin bond fatigue resistance and interfacial characteristics of universal adhesives in etch-and-rinse and self-etch modes. The two null hypotheses tested were as follows: (i) etching mode would make no differences to the bond fatigue resistance of universal adhesives to dentin, and (ii) there would be no differences in the interfacial characteristics of universal adhesive treated dentin.

Materials and Methods

1. Study materials

Five universal adhesives were used in this study: (i) Adhese Universal (AU, Ivoclar Vivadent, Schaan, Liechtenstein); (ii) All-Bond Universal (AB, Bisco, Schaumburg, IL, USA); (iii) Clearfil Universal Bond Quick (CU, Kuraray Noritake Dental), (iv) G-Premio Bond (GB, GC, Tokyo, Japan); and (v) Scotchbond Universal Adhesive (SU, 3M Oral Care). Ultra-Etch (Ultradent, South Jordan, UT, USA) was used as a 35% phosphoric acid etching agent, and Z100 Restorative (3M Oral Care) as the resin composite employed to make specimens (Table 1).

2. Specimen preparation

De-identified extracted human molar teeth were used in this study. The experimental protocol for using de-identified human molar teeth was reviewed and approved by the Ethics Committee for Human Studies of Nihon University School of Dentistry (No.2015-06) and Biomedical Institutional Review Board at Creighton University School of Dentistry (No.760765-1). Sectioned buccal and lingual halves of the teeth with the apical portions removed were mounted in 25-mm brass rings using an acrylic resin (Bosworth Fastray; Keystone Industries, Myerstown, PA, USA). Flat dentin surfaces were prepared on the mounted buccal and lingual surfaces by wet grinding using a gradually increasing sequence (#180-, #320-, #600-, #1,200-, #2,000-, and #4,000-grit) of silicon carbide (SiC) papers (Struers, Cleveland, OH, USA) in a grinder-polisher (Ecomet 4; Buehler, Lake Bluff, IL, USA). As the directionality of surface scratches created by the abrasives might have a substantial influence on the bond strength testing results, the surfaces were polished up to #4,000-grit to minimize this influence. These surfaces were then washed with water and dried using a dental three-way syringe at a distance of 5 cm above the surface and an air pressure of 0.3 MPa.

Thirty specimens were prepared for each of the adhesives for initial bond strength testing and 40 specimens were prepared for each of the adhesives for bond fatigue strength testing. In addition, 20 specimens were prepared for each of the adhesives for surface free-energy measurements. Half of the specimens for each of the adhesives were phosphoric acid etched for 15 s before application of the adhesive (etch-and-rinse mode), while the other half were not etched (self-etch mode). The specimens were prepared under ambient laboratory conditions of 23 ± 2°C and 50 ± 10% relative humidity.

3. Initial bond strength testing

Stainless steel (SUS304) rings with an inner diameter of 2.4 mm, an outer diameter of 4.8 mm, and a height of 2.6 mm were used to bond a resin composite to the dentin. The bonding side of the metal rings was treated with a 3% solution of paraffin in hexane. The dentin surfaces were treated with the universal adhesives in etch-and-rinse and self-etch mode according to the manufacturers' instructions. A custom fixture was used to position and hold the stainless steel rings over the bonding sites as the resin composite was placed into the rings using a condensing instrument. The resin composite was then light-cured for 40 s using a quartz-tungsten-halogen (QTH) curing unit (Spectrum 800 Curing Unit; Dentsply Caulk, Milford, DE, USA) set at light intensity of 800 mW/cm^2. The bonded specimens were then stored in 37ºC distilled water for 24 h before testing.

A chisel-shaped metal rod was used to apply the load to the metal rings (mold-enclosed method) immediately adjacent to the flat dentin surfaces. Fifteen specimens per group were loaded to failure using an all-electric dynamic test instrument (ElectroPuls E1000; Instron, Canton, MA, USA) with a crosshead speed of 1 mm/min. Initial shear bond strength (MPa) was calculated from the peak load at failure divided by the bonded surface area.

4. Bond fatigue strength testing

A staircase method was used to perform the bond fatigue strength tests using the all-electric dynamic test instrument. The fatigue load was applied to the metal rings using a sine wave at a frequency of 20 Hz for 50,000 cycles or until failure occurred. The initial peak load for bond fatigue strength testing for each of the adhesives was set at a level of approximately half of the initial bond strength for each group. The lower load limit was set at 0.4 N, and subsequent loading was adjusted upward or downward approximately 10% from the previous load depending on specimen survival or failure. This procedure was repeated for twenty specimens per group. The test specimens were immersed in room temperature water (23 ± 2°C) during this testing.

5. Surface free energy measurement

The dentin surfaces were prepared as described above. Each dentin surface was treated with universal adhesive in etch-and-rinse or self-etch mode in accordance with the manufacturers' instructions, and the uncured adhesive layer was removed by three alternating rinses using acetone and distilled water. The contact angles of the specimens were then measured to analyze the surface free-energy characteristics of each adhesive treated surface. Phosphoric acid etched and ground dentin surfaces were also measured. The surface free energy characteristics of the specimens were determined by measuring the contact angle formed with the surface by the three test liquids 1-bromonaphthalene, diiodomethane, and distilled water, each of which has known surface free-energy parameters. For each test liquid, the equilibrium contact angle (θ) was measured using the sessile drop method under ambient laboratory conditions, as described earlier, using a contact angle measurement apparatus (DM 500; Kyowa Interface Science, Saitama, Japan) for ten specimens per group. The apparatus was fitted with a charge-coupled device camera to enable automatic measurement. A standardized 1.0 µl drop of each test liquid was placed on the treated dentin surface, and a profile image was captured after 500 ms using the apparatus. Contact angles were then calculated using the $\theta/2$ method, using the built-in interface measurement and analysis system (FAMAS; Kyowa Interface Science). The surface free-energy (γ_S) and its parameters for the solids (γ_S^d, γ_S^p and γ_S^h) were calculated using the formulae which were described by Hata et al. (14), again using the built-in software (FAMAS).

6. Scanning Electron Microscopy (SEM) observation of bonding interface

Representative SEM micrographs of the resin-dentin interfaces for three specimens per group were obtained using field-emission SEM (ERA 8800FE; Elionix, Tokyo, Japan). A rectangular (4 × 2 × 1 mm) section of dentin was removed from the molars for the SEM observations of the bonding interface. The dentin surfaces were prepared as described above for specimen preparation. The dentin surfaces were treated with the adhesives according to the manufacturers' instructions in etch-and-rinse or self-etch mode, the resin composite placed, and then the resin composite was photo-cured for 40 s using the QTH curing unit from a standardized distance of 1 mm. For the resin composite/dentin interfaces, bonded specimens were embedded in epoxy resin (Epon 812; Nisshin EM, Tokyo, Japan) and then stored at 37°C for 24 h. They were then sectioned near the center of the bonded specimen and the surfaces of the cut halves polished with #180-, #320-, #600-, #1200-, #2,000- and #4,000-grit SiC paper using a grinder-polisher. Finally, the surfaces were polished with a soft cloth using 1.0 µm-grit diamond paste (DP-Paste; Struers, Ballerup, Denmark). SEM specimens of the resin-dentin interfaces were dehydrated by first immersing them in ascending concentrations of aqueous *tert*-butanol (50% for 20 min, 75% for 20 min, 95% for 20 min, and 100% for 2 h) and then transferred from the final 100% bath to a freeze drying apparatus (Model ID-3; Elionix) for 30 min. The polished surfaces were etched for 30 s using an argon ion-beam (Type EIS-200ER; Elionix) directed perpendicular to the surface at an accelerating voltage of 1.0 kV and ion current density of 0.4 mA/cm^2. The surfaces were then coated with a thin film of gold in a vacuum evaporator (Quick Coater Type SC-701; Sanyu Electron, Tokyo, Japan) and observed using field-emission SEM with an operating voltage of 10 kV.

7. Statistical analysis

Initial bond strength data were analyzed with two-way analysis of variance (ANOVA), using the factors adhesive and etching mode, followed by Tukey's post-hoc honestly significant difference (HSD) test. The bond fatigue strength data were analyzed using a modified *t*-test with a Bonferroni correction (custom program). The γ_S, γ_S^d, γ_S^p, and γ_S^h data were analyzed using one-way ANOVA along with Tukey's HSD test. All statistical analyses, apart from the bond fatigue strength data analysis, were conducted using statistical software (SPSS Statistics ver. 13; International Business Machines, Armonk, NY, USA) and the significance level was set at 0.05.

Results

1. Initial bond strength

The initial bond strengths of the universal adhesives in the etch-and-rinse and self-etch modes are shown in Table 2. The initial bond strengths of the universal adhesives were not influenced by the etching mode ($p > 0.05$), and the percentage differences of the initial bond strength between etch-and-rinse and self-etch modes were within 10% (3–10%). The initial bond strength was material-dependent regardless of etching mode. The initial bond strength of SU (39.3–41.5 MPa) was significantly higher than those of AU, AB, CU and GP (26.2–29.0 MPa).

2. Bond fatigue strength

The bond fatigue strengths for the universal adhesives in the etch-and-rinse and self-etch modes are shown in Table 3. The bond fatigue strengths of the universal adhesives were not influenced by the etching mode ($p > 0.05$), and the percentage differences of bond fatigue strength between etch-and-rinse and self-etch modes were less than 11% (5–11%). The bond fatigue strength was material-dependent regardless of the etching mode. The bond fatigue strength of SU (19.0–20.6 MPa) was significantly higher than those of AU, AB, CU and GB (12.3–14.4 MPa).

3. Surface free energy characteristics of adhesive treated dentin

The surface free energy and parameters of universal adhesive-treated dentin are shown in Table 4. The baseline in the etch-and-rinse mode exhibited a significantly lower γ_S and γ_S^h than that in the self-etch mode ($p < 0.05$). Changes in the γ_S^d and γ_S^p of universal adhesive-treated dentin were not influenced ($p > 0.05$) by the type of adhesive used, in contrast to γ_S^h which was observed to be influenced by the type of adhesive ($p < 0.05$).

4. SEM observation of bonding interface

Representative SEM images of the resin-dentin interface of etch-and-rinse and self-etch modes are shown in Figure 1. The thickness of the adhesive layer of universal adhesives was approximately 7–12 µm, and the resin-dentin interface of the tested adhesives showed excellent adaptation to dentin regardless of the etching mode. However, cracks were visible in the adhesive layer of the resin–dentin interface of GB in both etch-and-rinse and self-etch modes, but not in the other adhesives.

Discussion

Research into the application of compressive shear load in a conventional shear bond strength test using finite element stress analysis was previously conducted by Van Noort et al. (15) and has shown that the load is not uniformly distributed along the bonding interface. This was also confirmed in a recent study by Jin et al. (16). The conventional shear bond strength test has long been criticized as neither appropriate nor reliable for the measurement of "true" or "actual" shear bond strength at the bonding interface and may have been measuring "mixed" shear bond strength. In the modified shear bond strength test using dynamic loading in this study, the non-uniform stress distribution created by shear stress may be a more important issue than in the conventional shear bond strength test due to the repeated stress loading. The mold-enclosed format was utilized in this study, in which a stainless steel ring encloses the resin composite when both initial bond strength and bond fatigue strength are measured. It has been reported that with this method, non-uniform stress is significantly reduced, while the desired shear stress is maintained in shear bond strength tests, and finite element analysis suggests that this approach is more suitable for these measurements (16). Aside from minimizing radius effects at the bonding interface, the mold-enclosed method has the ability to reduce the load bearing on the resin composite itself, as the force is applied indirectly through the metal ring (17). Hence, the mold-enclosed method was chosen as a fitting approach for bond strength testing of universal adhesives.

In the present study, etching mode did not affect the bond fatigue resistance of universal adhesives to dentin, and thus the first null hypothesis was not rejected. A recent study (18), which used the same research design to study enamel bonding, reported that the bond fatigue resistance of universal adhesives was significantly higher in etch-and-rinse mode than in self-etch mode. The results of the present and previous studies suggested that the use of etch-and-rinse or selective etching modes with universal adhesives is more effective from the bond fatigue resistance perspective, in agreement with a systematic review of earlier laboratory bond strength evaluations (19).

The results for the interfacial characteristics showed that the baseline γs, γs^d, γs^p, and γs^h of dentin in the etch-and-rinse mode were different and significantly below those in the self-etch mode. This means that phosphoric acid etched dentin has lower wettability and degree of polarization and is less hydrophilic than ground dentin. Tay et al. (20) have repeatedly reported that dehydration of demineralized dentin results in osmosis of water content from deeper dentin, leading to weaker bonding due to osmotic blisters and hydrolysis of the adhesive itself. However, bonding strategy did not influence the dentin bond fatigue resistance of universal adhesives. In the SEM observation of adhesive interfaces in etch-and-rinse mode, deeper penetration of adhesives into the dentinal tubules was observed due to the removal of the smear layer and opening of the dentinal tubules by phosphoric acid etching, despite the lower wettability of the dentin. These deeper resin tags, which were perpendicular to shear stress, may have contributed to the resistance of adhesive interfaces to cyclic fatigue. In addition, bond fatigue testing was done with 20 Hz frequency and a cycling period of 50,000 cycles, in approximately 40 min, which is not very long. Hence, the influence of osmosis of water content from deeper dentin, resulting in osmotic blisters and hydrolysis of adhesive, might be reduced due to the shorter period of testing compared to tests using long-term water storage or thermal cycling.

However, the bond fatigue resistance of universal adhesives showed a dependence on the material. The bond fatigue resistance of SU was significantly higher than that of the other adhesives, regardless of etching mode. Although no clear relationship between bond fatigue resistance and interfacial characteristics was seen, the measurements of the interfacial characteristics of adhesive-treated dentin may make differences among the adhesives tested clearer. For universal adhesives, 10-MDP is a key technological factor for chemical bonding with dentin substrate, regardless of the bonding strategies employed. The adhesion-decalcification concept (21) claimed that the functional group of 10-MDP interacts ionically with calcium in dentin and forms a chemically bonded 10-MDP-calcium salt-layered structure on the dentin surface. As the long carbonyl chain of 10-MDP is relatively hydrophobic (22), a hydrophobic layer would cover the dentin surface due to the 10-MDP-calcium salt-layered structure. A greater extent of chemical bonding to the dentin surface should therefore create a more hydrophobic surface. The results for γs and γs^h on universal adhesive-treated dentin surfaces depended on the material regardless of etching mode, unlike γs^d and γs^p. Thus, the second null hypothesis was partially rejected. Further, γs and γs^h on GB- and SU-treated surfaces in both etching groups were significantly lower than those of AU-, AB- and CU-treated surfaces. A previous study (13) reported that interfacial characteristics were influenced by the reactions between acidic functional monomers and calcium in tooth substrates; thus these results are consistent with previous studies. Furthermore, the interfacial characteristics of adhesive-treated surfaces for all tested adhesives decreased compared to those of the baseline regardless of etching mode, suggesting that the adhesives were capable of forming chemical bonds to both etched and ground dentin, creating a surface layer with distinct chemical properties.

On the other hand, even though GB-treated dentin surface had a lower γs, similar to SU, GB showed a lesser bond fatigue resistance than SU, and there was no significant difference from AU, AB or CU. SEM observations revealed clear qualitative differences in the adhesive layers. Cracks and defects were observed in the adhesive layer of GB-dentin interfaces, in both etching modes, but not in the other adhesives tested. A previous study of the water content of universal adhesives (23) reported that GB contains approximately 25% water, a higher proportion than other universal adhesives, where the proportion can be as low as 3%, in AU. SU has approximately half the proportion of water found in GB, at 10 to 15%. The extra water in GB is present to enhance its demineralization of tooth substrates, but might lead to weakness caused by cracks, especially if there is residual water in the cured adhesive layer. This partially explains why the bond fatigue resistance of GB was lower than that of SU and similar to the other adhesives, even though GB-treated dentin showed lower surface free energies.

Taken together, these results suggested that the bonding mechanisms in universal adhesives may vary substantially between adhesives, and between etching mode for the same adhesive. A better understanding of these mechanisms may allow significant improvements to be made to the bonding performance of universal adhesives, and thus is an important topic for further research.

Conclusion

1. The initial bond strengths were not influenced by the etching mode, and the percentage difference of the initial bond strength between the two modes was within 10%. The initial bond strengths were material-dependent regardless of etching mode.
2. Bond fatigue strengths were not influenced by the etching mode, and percentage differences of bond fatigue strength between the two modes were less than 11%. The bond fatigue strengths were material-dependent regardless of the etching mode.
3. The baseline in the etch-and-rinse mode exhibited a significantly lower γs and γs^h than that in the self-etch mode. The γs and γs^h of universal adhesive-treated dentin were decreased unlike γs^d and γs^p, regardless of etching mode.
4. The adhesive layer thickness was different depending on the adhesive, and the adaptation of the interface to dentin was excellent for both etching modes. Cracks were visible in the adhesive layer of the resin/dentin interface of GB in both etching groups, but not in the other adhesives tested.

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Table 1: Materials used in this study

Material (Lot No.)	Type of material (Code)	Main component (pH)	Manufacturer
Adhese Universal (164453)	Universal adhesive (AU)	Bis-GMA, HEMA, MDP, MCAP, decandiol dimethacrylate, dimethacrylate, ethanol, water, initiator, stabilizers, silicon dioxide (2.5)	Ivoclar Vivadent, Schaan, Liechtenstein
All-Bond Universal (1300008503)	Universal adhesive (AB)	Bis-GMA, HEMA, MDP, ethanol, water, initiators (3.2)	Bisco, Schaumburg, IL, USA
Clearfil Universal Bond Quick (1L0003)	Universal adhesive (CU)	Bis-GMA, HEMA, MDP, hydrophilic amide monomer, ethanol, water, initiators, silica, silane coupling agent (2.3)	Kuraray Noritake Dental, Tokyo, Japan
G-Premio Bond (1603091)	Universal adhesive (GB)	MDP, 4-MET, MEPS, methacrylate monomer, acetone, water, initiator, silica (1.5)	GC, Tokyo, Japan
Scotchbond Universal Adhesive (617265)	Universal adhesive (SU)	Bis-GMA, HEMA, decamethylene dimethacrylate, ethyl methacryalate, propenoic acid, methyl-reaction products with decanediol and phosphorous oxide, copolymer of acrylic and itaconic acid, dimethylaminobenzoate, methyl ethyl ketone, ethanol, water, silane treated silica, initiator (2.7)	3M Oral Care, St. Paul, MN, USA
Ultra-Etch (G019)	Etching agent	35% phosphoric acid, glycol, cobalt aluminate blue spinel	Ultradent Products, South Jordan, UT, USA
Z100 (1312131)	Resin composite	Bis-GMA, TEGDMA, silane treated ceramic, benzotriazolyl methylphenol	3M Oral Care

Bis-GMA: 2,2-bis[p-(2-hydroxy-3-methacryloxy propoxy)phenyl]propane, HEMA: 2-hydroxyethyl methacrylate, MDP: 10-methacryloyloxydecyl di-hydrogen phosphate, MCAP: methacrylated carboxylic acid polymer, 4-MET: 4-methacryloyloxyethyl trimellitate, MEPS: methacryloyloxyalkyl thiophosphate methylmethacrylate, TEGDMA: triethylene glycol dimethacrylate

Table 2: Initial shear bond strength of universal adhesives to dentin using etch-and-rinse and self-etch modes

Code	Etch-and-rinse mode	Self-etch mode
AU	29.0 (4.2)^{a,A}	26.4 (5.5)^{a,A}
AB	27.2 (3.8)^{a,A}	27.9 (3.7)^{a,A}
CU	27.3 (4.2)^{a,A}	28.1 (4.2)^{a,A}
GB	26.2 (5.5)^{a,A}	27.5 (3.5)^{a,A}
SU	39.3 (4.6)^{b,A}	41.5 (4.5)^{b,A}

Unit: MPa. Values in parenthesis are standard deviations. Same small letters in same individual column indicate no significant difference ($p > 0.05$). Same capital letters within individual rows indicate no significant difference ($p > 0.05$).

Table 3: Bond fatigue strength of universal adhesives to dentin using etch-and-rinse and self-etch modes

Code	Etch-and-rinse mode	Self-etch mode
AU	13.7 (1.9)^{a,A}	12.3 (3.3)^{a,A}
AB	13.1 (2.5)^{a,A}	13.8 (2.4)^{a,A}
CU	13.9 (1.9)^{a,A}	14.4 (1.7)^{a,A}
GB	13.0 (2.2)^{a,A}	14.3 (2.8)^{a,A}
SU	19.0 (3.1)^{b,A}	20.6 (2.8)^{b,A}

Unit: MPa. Values in parenthesis are standard deviations. Same small letters in same individual column indicate no significant difference ($p > 0.05$). Same capital letters within individual rows indicate no significant difference ($p > 0.05$).

Table 4: Surface free energy characteristics of universal adhesive treated dentin using etch-and-rinse and self-etch modes

Code	Etch-and-rinse mode				Self-etch mode			
	γ_s	γ_s^d	γ_s^p	γ_s^h	γ_s	γ_s^d	γ_s^p	γ_s^h
baseline	41.1 (2.5)^{a}	37.8 (1.5)^{a}	1.1 (1.6)^{a}	2.2 (1.1)^{a}	68.8 (3.6)^{a}	41.0 (1.4)^{a}	2.8 (2.3)^{a}	25.0 (2.4)^{a}
AU	39.6 (2.3)^{a,b}	37.0 (1.7)^{a}	1.4 (2.1)^{a}	1.2 (0.9)^{b}	65.5 (3.7)^{b}	40.0 (1.4)^{a}	2.6 (2.2)^{a}	22.9 (2.6)^{b}
AB	39.0 (2.2)^{a,b}	37.1 (0.4)^{a}	1.0 (1.9)^{a}	0.9 (2.0)^{b}	63.8 (3.0)^{b}	40.1 (0.4)^{a}	2.4 (1.7)^{a}	21.3 (2.2)^{b}
CU	39.2 (2.4)^{a,b}	37.2 (0.8)^{a}	1.1 (1.8)^{a}	0.9 (2.1)^{b}	64.1 (2.9)^{b}	40.2 (1.1)^{a}	2.5 (1.5)^{a}	21.4 (2.0)^{b}
GB	37.7 (1.9)^{b}	37.2 (1.6)^{a}	0.4 (1.0)^{a}	0.1 (0.3)^{c}	61.1 (3.0)^{b,c}	40.3 (1.5)^{a}	2.6 (2.1)^{a}	18.2 (3.1)^{b,c}
SU	38.0 (2.2)^{b}	37.1 (1.6)^{a}	0.7 (1.5)^{a}	0.2 (0.4)^{d}	62.0 (3.7)^{b,c}	40.8 (1.5)^{a}	1.5 (2.0)^{a}	19.7 (2.7)^{b,c}

Unit: mN/m. γ_s, surface free-energy; γ_s^d dispersion force; γ_s^h, hydrogen-bonding force; γ_s^p, polar force. The same letters in a column indicate no statistically significant difference ($p > 0.05$).

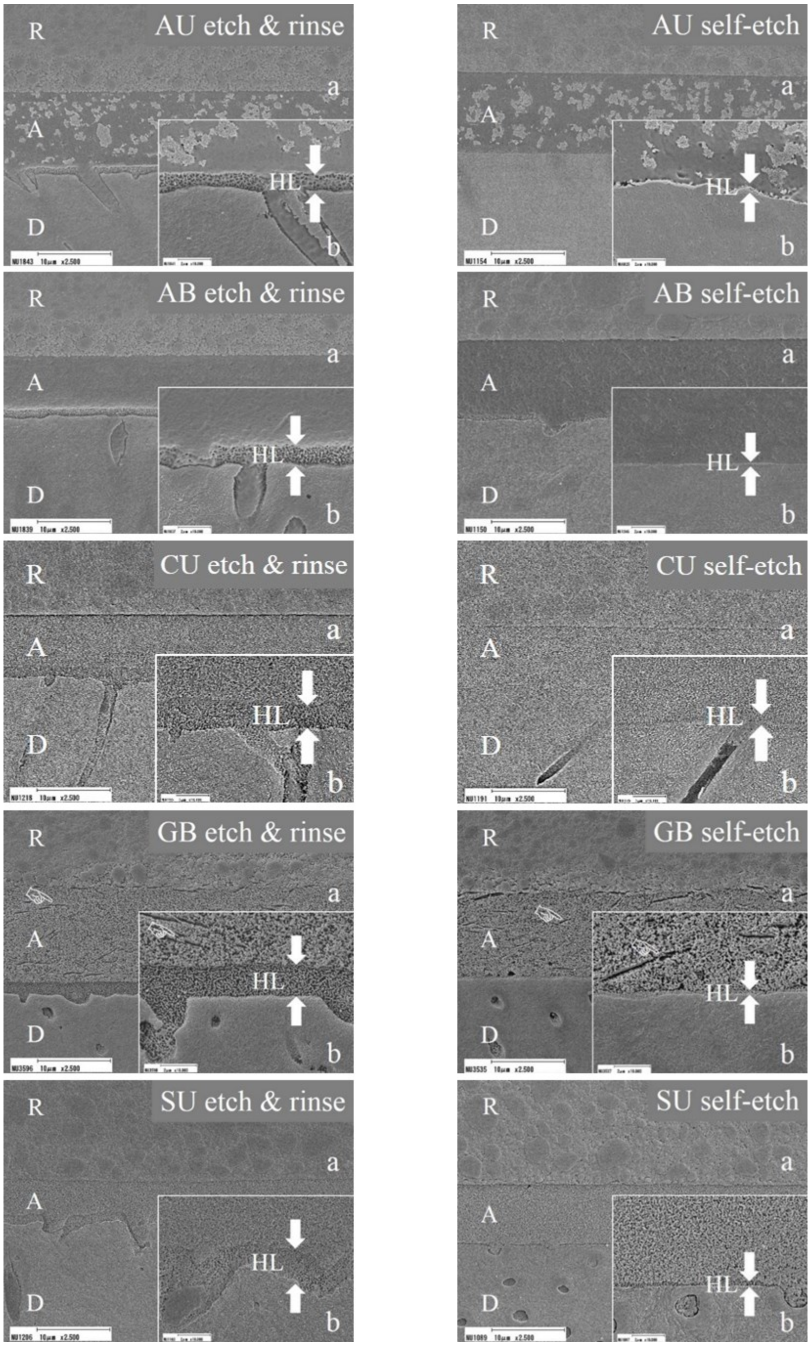

Figure 1: Scanning electron microscopy images of the resin-dentin interface at (a) x2,500 and (b) x10,000 magnifications. A, Adhesive; D, dentin; HL, hybrid layer; R, resin composite; AU, Adhese Universal; AB, All-Bond Universal; CU, Clearfil Universal Bond Quick, GB, G-Premio Bond; SU, Scotchbond Universal Adhesive.