Original article

Etch-and-rinse vs self-etch mode for dentin bonding effectiveness of universal adhesivesKabun Yamauchi¹⁾, Akimasa Tsujimoto¹⁾, Carlos A. Jurado^{2,3)}, Yusuke Shimatani¹⁾, Yuko Nagura¹⁾, Toshiki Takamizawa¹⁾, Wayne W. Barkmeier⁴⁾, Mark A. Latta⁴⁾, and Masashi Miyazaki¹⁾¹⁾ Department of Operative Dentistry, Nihon University School of Dentistry, Tokyo, Japan²⁾ Department of Restorative Dentistry, Oregon Health and Science University School of Dentistry, Portland, OR, USA³⁾ Department of Digital Dentistry, A.T. Still University Arizona School of Dentistry, Mesa, AZ, USA⁴⁾ Department of General Dentistry, Creighton University School of Dentistry, Omaha, NE, USA

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Abstract: In this study, dentin bond fatigue resistance and interfacial science characteristics of universal adhesives through etch-and-rinse and self-etch modes were investigated. Resin composite was bonded to human dentin with four universal adhesives, namely, Adhese Universal, All-Bond Universal, G-Premio Bond, and Scotchbond Universal Adhesive. The initial bond strengths, bond fatigue strengths, and interfacial science characteristics of the universal adhesives with dentin through etch-and-rinse and self-etch modes were determined. Bond fatigue resistance (initial bond strength and bond fatigue strength) of universal adhesives in etch-and-rinse mode showed no significant difference in contrast to that in self-etch mode and was material-dependent regardless of the etching mode. Although phosphoric acid conditioning of dentin did not have a strong impact on the bond fatigue resistance, surface free energy and parameters of dentin were significantly decreased by etching and by application of universal adhesives regardless of etching mode. Changes in γ_s and γ_s^h for when universal adhesive was applied to etched and ground dentin were significantly different depending on the adhesive. The results suggest that bonding performance of universal adhesives was effective in both etching modes; however, bonding mechanisms may be different for each.

Keywords; light-curing of dental adhesives, dental debonding, dental etching, dental restoration failure, dental stress analysis

Introduction

Universal adhesives have become a trend in dentistry because they can be used in either etch-and-rinse or self-etch mode, thanks to their reduced number of application steps and flexibility [1]. With the expiration of the patent for the 10-MDP (10-methacryloyloxydecyl dihydrogen phosphate) from Kuraray Noritake Dental in 2003, manufacturers began exploring the usage of 10-MDP and other phosphoric acid ester for 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 which could be of use with resin luting cements [2], various substrates without surface treatment [3], reduced treatment times [4], or various surface moisture of enamel and dentin surfaces [5] were introduced in the market. Despite this exciting flexibility, some reports suggest that the bond durability of numerous universal adhesives is inferior to that of the two-step self-etch adhesives. With enamel, this has been seen in etch-and-rinse and self-etch modes [6,7] and dentin in self-etch mode [8]. Nevertheless, flexibility of universal adhesives has enjoyed increasingly popularity in the clinical setting [9], hence the 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 by a collaborative effort between the Nihon University School of Dentistry (NU, Tokyo, Japan) and Creighton University School of Dentistry (CU, Omaha, NE, USA). Development started with Erickson et al. [10,11] and was refined by researchers from

NU and CU (Barkmeier et al. [12], Takamizawa et al. [13], and Tsujimoto et al. [14]). The appropriate frequency [13,15], number of cycles [14], and analysis are now well-established. However, at the present time, research comparing the bonding performance of universal adhesives using this bond fatigue test for either etch-and-rinse or self-etch mode has been limited.

Tian et al. [16] reported that the chemical interaction of 10-MDP with dentin is vital in obtaining durable bonds. Inoue et al. [17] 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 strategies employed, findings on the chemical bonding interactions of universal adhesive to ground and etched dentin may have been disparate. In addition, further assessment on the changes of energetic characteristics of dentin surfaces treated by universal adhesives in the two different modes, when added to the bond fatigue resistance analysis, may provide an explanation for the discrepancy between laboratory and clinical results.

The aim of this laboratory study was to assess dentin bond fatigue resistance and interfacial science 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 science characteristics of universal adhesive-treated dentin.

Materials and Methods**Study materials**

Four 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) G-Premio Bond (GB, GC, Tokyo, Japan), and (iv) Scotchbond Universal Adhesive (SU, 3M Oral Care). Ultra-Etch (Ultradent, South Jordan, UT, USA) was used as a 35% phosphoric acid pre-etching agent and Z100 Restorative (3M Oral Care) as the resin composite employed to make specimens. Table 1 lists the tested materials and their lot numbers and components.

Specimen preparation

This study used de-identified extracted human molar teeth. To comply with research ethics, the protocol was reviewed and approved by the Ethics Committee for Human Studies of NU (#2015-06) and Biomedical Institutional Review Board at CU (#760765-1). Preparation of the teeth was done in the manner described in the study by Nagura et al. [18], except that a flat dentin surface, rather than enamel surface, was prepared. Specimens with and without etching were prepared for each adhesive, giving a total of eight experimental groups for each of the procedures performed (Table 2).

Initial bond strength testing

Initial bond strength testing was carried out as described by Nagura et al. [18].

Bond fatigue strength testing

Bond fatigue strength testing was carried out as described by Nagura et al. [18].

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Table 1 Materials used in this study

Material	Type of material (Code)	Main components (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
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 methacrylate, 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)	Pre-etching agent	35% phosphoric acid, glycol, cobalt aluminate blue spinel	Ultradent Products, South Jordan, UT, USA
Z100 (1312131)	Resin composite (Z100)	Bis-GMA, TEGDMA, silane-treated ceramic, benzotriazolyl methylphenol	3M ESPE

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

Table 2 Application protocols used for phosphoric acid etching and for adhesive application

Etching mode	Phosphoric acid pre-etching protocol
Etch-and-rinse mode	Dentin surface was conditioned with phosphoric acid for 15 s. Conditioned surface was rinsed with water for 15 s (three-way dental syringe) and air-dried
Self-etch mode	Phosphoric acid etching was not performed
Adhesive	Adhesive application protocol
AU	Adhesive applied to air-dried tooth surface with rubbing action for 20 s and then medium air pressure applied to surface for 5 s. Photo-polymerized for 10 s
AB	Adhesive applied to enamel surface with rubbing action for 10-15 s and then medium air pressure applied to surface for 10 s. Photo-cured for 10 s
GB	Adhesive applied to air-dried tooth surface for 10 s and then maximum air pressure applied to surface for 5 s. Photo-cured for 10 s
SU	Adhesive applied to air-dried tooth surface with rubbing action for 20 s and then medium air pressure applied to surface for 5 s. Photo-cured for 10 s

Fig. 1 Illustration of the apparatus for contact angle measurement

Failure mode of specimens

Failure mode was assessed as described by Nagura et al. [18].

Interfacial science characteristics measurement

Interfacial science characteristics were measured as described by Nagura et al. [18]. The procedure is depicted in Fig. 1.

Scanning electron microscopy (SEM) observation of bonding interface

SEM observations were conducted as described by Nagura et al. [18].

Statistical analysis

Statistical analysis was conducted as described by Nagura et al. [18], with

the exception that Fisher's exact test was not used in this study.

Results

Initial bond strength

In Table 3 are the initial bond strengths of the universal adhesives in both etching mode groups. The initial bond strengths showed no influence ($P > 0.05$) by the etching mode, and the percentage difference of the initial bond strength between the two modes was within 10% (3-10%). The initial bond strength was material-dependent regardless of etching mode.

Bond fatigue strength

Table 4 shows the bond fatigue strengths in both etching mode groups.

Table 3 Initial shear bond strength of universal adhesives to dentin using etch-and-rinse and self-etch modes

Adhesive	Etch-and-rinse mode	Self-etch mode	Percentage difference
AU	29.0 (4.2) ^{a,A}	26.4 (5.5) ^{a,A}	10%
AB	27.2 (3.8) ^{a,A}	27.9 (3.7) ^{a,A}	3%
GB	26.2 (5.5) ^{a,A}	27.5 (3.5) ^{a,A}	5%
SU	39.3 (4.6) ^{b,A}	41.5 (4.5) ^{b,A}	6%

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

Adhesive	Etch-and-rinse mode	Self-etch mode	Percentage difference
AU	13.7 (1.9) ^{a,A}	12.3 (3.3) ^{a,A}	11%
AB	13.1 (2.5) ^{a,A}	13.8 (2.4) ^{a,A}	5%
GB	13.0 (2.2) ^{a,A}	14.3 (2.8) ^{a,A}	10%
SU	19.0 (3.1) ^{b,A}	20.6 (2.8) ^{b,A}	8%

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 7 Surface free energy characteristics of universal adhesive-treated dentin using total-etch and self-etch modes

Adhesive	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
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 , total free energy; γ_s^d , dispersion force; γ_s^h , hydrogen-bonding force; γ_s^p , lar force. The same letters in a column indicate no statistically significant difference ($P > 0.05$).

Bond fatigue strengths were not influenced ($P > 0.05$) by the etching mode, and percentage differences of bond fatigue strength between the two modes were less than 11% (5-11%). The bond fatigue strength was material-dependent regardless of the etching mode.

Failure mode analysis

The failure mode analysis for initial bond strength is shown in Table 5, while that for bond fatigue strength testing is in Table 6. Most of the failure modes were adhesive failure, and neither type of adhesive nor the etching mode made a difference.

Interfacial science characteristics of adhesive-treated dentin

Table 7 shows the interfacial science characteristics of dentin treated with the universal adhesives. The baseline in the etch-and-rinse group exhibited a significantly lower γ_s and γ_s^h ($P < 0.05$) than that in the self-etch group. Changes in γ_s^d and γ_s^p were found to have not been influenced ($P > 0.05$) by the type of adhesive used, in contrast to changes in γ_s^h which were observed to be influenced by type of adhesive ($P < 0.05$).

SEM observation of bonding interface

Figure 2 shows representative SEM images of resin-dentin interfaces from both etching groups. The adhesive layer was approximately 7-12 μm thick, and the adaptation of the interface to dentin was excellent for both etching modes. However, cracks were visible in the adhesive layer of the resin-dentin interface of GB in both groups, 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. [19] and has shown lack of uniform distributed load along the bonding interface. This was also confirmed through the recent study reported by Jin et al. [20]. 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.

Table 5 Failure mode analysis of debonded specimens after initial bond strength tests

Adhesive	Etch-and-rinse mode	Self-etch mode
AU	[86/0/7/7]	[100/0/0/0]
AB	[100/0/0/0]	[100/0/0/0]
GB	[86/0/7/7]	[100/0/0/0]
SU	[86/0/14/0]	[100/0/0/0]

Percentage of failure mode [adhesive failure/cohesive failure in resin/cohesive failure in enamel/mixed failure]. Same small letters in same column indicate no significant difference ($P > 0.05$). Same capital letters within individual rows indicate no significant difference ($P > 0.05$).

Table 6 Failure mode analysis of debonded specimens after shear fatigue strength tests

Adhesive	Etch-and-rinse mode	Self-etch mode
AU	[90/0/5/5]	[100/0/0/0]
AB	[90/0/10/0]	[100/0/0/0]
GB	[80/0/10/10] ^a	[90/0/10/0]
SU	[60/10/20/10]	[100/0/0/0]

Percentage of failure mode [adhesive failure/cohesive failure in resin/cohesive failure in enamel/mixed failure]. Same small letters in same column indicate no significant difference ($P > 0.05$). Same capital letters within individual rows indicate no significant difference ($P > 0.05$).

Especially in the modified shear bond strength test using dynamic loading in this study, a non-uniform stress distribution which is made by shear stress may be a debatable larger issue than in the conventional shear bond strength test due to the repeated stress loading. A mold-enclosed format was utilized in this study in which a stainless steel metal ring encloses the resin composite when both initial bond and fatigue strength are measured. It has been reported that with utilization of this method, tensile stress is significantly reduced while desired shear stress is maintained in shear bond strength tests, and finite element analysis suggests that it is more suitable for these measurements [20]. Aside from minimizing radius effects at the bonding interface, the mold-enclosed method has the ability to reduce load bearing on the resin composite itself, as the force is applied indirectly through the metal ring [21,22]. Barkmeier et al. [12] previously reported that with metal ring-enclosed method employed, shear bond strengths were generally 20% higher than the results achieved with use of Ultradent shear bond strength method. Hence, the mold-enclosed method was chosen as a fitting tool for bond fatigue resistance analysis of universal adhesives.

The results of the present study showed that etching mode did not affect the bond fatigue resistance of universal adhesives to dentin. However, the first null hypothesis that etching mode would make no difference to the bond fatigue resistance of universal adhesives to dentin was not rejected. A recent study [18], which uses the same research design to study bonding to enamel, has reported that the bond fatigue resistance of universal adhesives was significantly higher in etch-and-rinse 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 perspectives in laboratory, in agreement with the systematic review of earlier laboratory bond strength evaluations [23].

Results for the interfacial science characteristic evaluation show the baseline γ_s , γ_s^d , γ_s^p , and γ_s^h of dentin in the etch-and-rinse group (phosphoric acid etched dentin) were different and significantly below those in the self-etch group (ground dentin), where γ_s is the total surface free energy; γ_s^d is the dispersion force of the solid surface which is related to its wettability [17]; γ_s^p is the polarity force, which represents degree of polarization; and γ_s^h is the hydrogen-bonding force, which indicates

Fig. 2 Typical scanning electron microscopy images of the resin-dentin interface of etch-and-rinse and self-etch groups at (a) 2,500 $\times$ magnification and (b) 10,000 $\times$ magnification. A, Adhesive; D, dentin; HL, hybrid layer; R, resin composite; AU, Adhese Universal; AB, All-Bond Universal; GB, G-Premio Bond; SU, Scotchbond Universal Adhesive

the hydrophilicity of the surface [24]. This means that phosphoric acid etched dentin has lower wettability and degree of polarization and is less hydrophilic compared to ground dentin. Tay et al. [25,26] 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 bond fatigue resistance of these adhesives to dentin. In the SEM observation of adhesive interfaces in etch-and-rinse mode, the 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 higher resistance of adhesive interfaces to cyclic fatigue. In addition, fatigue testing was done with 20 Hz frequency and a cycling period of 50,000 cycles, in approximately 40 minutes, which is not very long. Hence, influence of osmosis of water content from deeper dentin, resulting in osmotic blisters and hydrolysis of adhesive, might be less 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 the other adhesives, regardless of etching mode. Although no clear relationship between bond fatigue resistance and interfacial science characteristics was seen, the measurements of interfacial science 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 tooth substrates, regardless of the bonding strategies employed [16]. The adhesion-decalcification concept [27] claims 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 [28], 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 γ_s and γ_s^h on universal adhesive-treated dentin surfaces depended on the material regardless of

etching mode, unlike γ_s^d and γ_s^p . Thus, the second null hypothesis that there would be no differences in the interfacial science characteristics of universal adhesive-treated dentin was rejected. Further, γ_s and γ_s^h on GB- and SU-treated surfaces in both etching groups were significantly lower than those of AU and AB. Previous studies [16,23,24] reported that interfacial science characteristics were influenced by the reaction between acidic functional monomers and calcium in tooth substrates; thus these results were consistent with previous studies. Furthermore, the interfacial science 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 unetched dentin, creating a surface layer with distinct chemical properties.

On the other hand, even though GB-treated dentin had a lower γ_s , similar to SU, GB showed a lesser bond fatigue durability than SU, and there was no significant difference from AU or AB. In 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 [29] 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 (i.e., 10 to 15%). The extra water in GB is present to enhance its demineralization of tooth substrates. The higher level of water content in GB might lead to weakness caused by cracks, especially 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.

GB also has a distinctive composition: The three other adhesives used in the study contained a single functional monomer, 10-MDP, while GB on the other hand contains several acidic functional monomers, which include 10-MDP, 4-methacryloxyethyl trimellitic acid (4-MET), and 10-methacryloyloxydecyl dihydrogen thiophosphate (10-MDTP). The bonding mechanisms of 4-MET and 10-MDP to dentin have been reported to be similar [30], creating a hydrophobic surface. Given this, it is reasonable to suppose that multiple monomers might contribute to lower γ_s and γ_s^h values of GB-treated dentin. Conversely, a previous study has reported that 4-MET has substantially lower chemical bonding potential than 10-MDP [31]. If 4-MET makes a substantial contribution to reducing γ_s and γ_s^h , but a smaller contribution to bond durability, this may also help to explain why GB had a lower bond fatigue resistance than SU despite the similar interfacial science characteristics results.

Taken together, these results suggest that the bonding mechanism in universal adhesives may vary substantially between adhesives and between application modes 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 is thus an important topic for further research.

Considering the results of present and previous studies which assessed enamel and dentin bond fatigue resistance with universal adhesives, universal adhesives used in etch-and-rinse mode may have better bond fatigue resistance because the etch-and-rinse mode leads to higher enamel bonding than self-etch mode and does not make a significant difference to dentin bond fatigue resistance. However, clinical studies [6,7] found there was no statistical difference among bonding strategies when a universal adhesive was used and that etching had only a minor effect with universal adhesives. These results may indicate that the observed increase of enamel bond fatigue resistance in etch-and-rinse mode fails to make an important clinical difference, most likely because dentin bond fatigue resistance has a much stronger influence. Research to clarify the relationship between bond fatigue resistance and clinical results would make a valuable contribution to the field.

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Conflict of Interest

The authors of this manuscript certify that they have no proprietary, finan-

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