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Effect of Oxygen Inhibition Layer of Universal Adhesives on Enamel Bond Fatigue Durability and Interfacial Characteristics With Different Etching Modes

H Ouchi • A Tsujimoto • K Nojiri • K Hirai
T Takamizawa • WW Barkmeier • MA Latta • M Miyazaki

Clinical Relevance

The oxygen inhibition layer of universal adhesives does not impair the enamel bond fatigue durability and interfacial characteristics of the adhesive regardless of etching mode, making it unnecessary for clinicians to consider it a cause for concern.

SUMMARY

Objective: The purpose of this study was to evaluate the effect of the oxygen inhibition layer of universal adhesive on enamel bond fatigue durability and interfacial characteristics with different etching modes.

Methods: The three universal adhesives used were Scotchbond Universal Adhesive (3M ESPE, St Paul, MN, USA), Adhese Universal (Ivoclar Vivadent, Schaan, Lichtenstein), and G-Premio Bond (GC, Tokyo, Japan). The initial shear bond strength and shear fatigue strength to enamel was determined in the presence and absence of the oxygen inhibition layer, with and without phosphoric acid pre-etching. The water contact angle was also measured in all groups using the sessile drop method.

Results: The enamel bonding specimens with an oxygen inhibition layer showed significant-

Hajime Ouchi, DDS, Operative Dentistry, Nihon University School of Dentistry, Tokyo, Japan

*Akimasa Tsujimoto, DDS, PhD, Operative Dentistry, Nihon University School of Dentistry, Tokyo, Japan

Kie Nojiri, DDS, PhD, Operative Dentistry, Nihon University School of Dentistry, Tokyo, Japan

Kazutaka Hirai, DDS, Operative Dentistry, Nihon University School of Dentistry, Tokyo, Japan

Toshiki Takamizawa, DDS, PhD, Operative Dentistry, Nihon University School of Dentistry, Tokyo, Japan

Wayne W Barkmeier, DDS, MS, General Dentistry, Creighton University, Omaha, NE, USA

Mark A Latta, DMD, MS, General Dentistry, Creighton University, Omaha, NE, USA

Masashi Miyazaki, DDS, PhD, Operative Dentistry, Nihon University School of Dentistry, Tokyo, Japan

*Corresponding author: 1-8-13, Kanda-Surugadai, Chiyoda-ku, Tokyo, 101-8310, Japan; e-mail: tsujimoto.akimasa@nihon-u.ac.jp

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ly higher ($p<0.05$) initial shear bond strengths and shear fatigue strengths than those without, regardless of the adhesive type and etching mode. Moreover, the water contact angles on the specimens with an oxygen inhibition layer were significantly lower ($p<0.05$) than on those without, regardless of etching mode.

Conclusion: The results of this study suggest that the oxygen inhibition layer of universal adhesives significantly increases the enamel bond fatigue durability and greatly changes interfacial characteristics, suggesting that the bond fatigue durability and interfacial characteristics of these adhesives strongly rely on its presence.

INTRODUCTION

The polymerization reaction in adhesive systems induced by photo-curing leads to the decomposition of camphorquinone and tertiary amine, resulting in the generation of reactive free radicals, which are able to add to the double bonds of resin monomers.[1] However, oxygen has a greater ability to react with the propagating free radicals than the monomer molecule, oxidizing them into peroxy radicals, which have relatively low reactivity toward the monomer and form peroxides, terminating polymerization if they do react.[2] This leads to retardation or inhibition of the free radical polymerization reaction[3] and consequently to the formation of an oxygen inhibition layer on the superficial surface of photo-cured resin-based materials when these are polymerized in the presence of air.[4]

The evidence concerning the role of the oxygen inhibition layer of adhesives is still controversial, with some studies reporting that the presence of an oxygen inhibition layer is necessary for bonding the adhesive to resin composite[1,5] and others reporting that this layer has no significant effect[6-8] or a negative effect[9-11] on bonding. Therefore, further studies are necessary in order to elucidate its effects on bond durability.

The development of next-generation adhesive systems has aimed to reduce technique sensitivity and the number of clinical steps,[12] leading to time-saving options such as single-step self-etch adhesives.[13] In conjunction with this trend, universal adhesives have been introduced to the profession.[14] Universal adhesives can be used with any of the total-etch, self-etch, or selective-etch techniques, and can bond to various substrates other than tooth substrates.[15] The increasing popularity of these adhesives, due to their versatility, has called into question the role of the oxygen inhibition layer in bonding.

Tsujimoto and others[16] have reported that the interfacial characteristics of the oxygen inhibition layer differ with the type of adhesive system. Its characteristics in photo-cured resin-based materials are influenced by several factors, including monomer chemistry,[4,8] filler morphology and temperature,[17,18] radical concentration,[2] and the rate of oxygen consumption.[19] Therefore, it is possible that the interfacial characteristics of the oxygen inhibition layer of universal adhesives may differ from those of other adhesive systems. These characteristics are often examined using water as a probe liquid and provide information on the suitability of a surface for bonding,[20] which affects the strength of the bond across the interface.[21] Thus, analyzing the interfacial characteristics of universal adhesives in terms of water contact angles may provide insights into the influence of the oxygen inhibition layer on their bond durability.

The purpose of this laboratory study was to investigate the effect of the oxygen inhibition layer of universal adhesives on enamel bond fatigue durability and interfacial characteristics. The null hypothesis tested posited that the enamel bond fatigue durability and interfacial characteristics of universal adhesives would not be influenced by the presence or absence of the oxygen inhibition layer.

METHODS AND MATERIALS

Study Materials

The three universal adhesives used were Scotchbond Universal Adhesive (SU; 3M ESPE, St Paul, MN, USA), Adhese Universal (AU; Ivoclar Vivadent, Schaan, Lichtenstein), and G-Premio Bond (GB; GC, Tokyo, Japan). Ultra-Etch (Ultradent Product, South Jordan, UT, USA) was used as a phosphoric acid pre-etching agent for bonding to enamel, and Z100 Restorative (3M ESPE) was used as the resin composite for the bonding procedures. The lot numbers and compositions of the materials used are listed in Table 1.

Initial Shear Bond Strength Tests

This study used extracted noncarious deidentified human molars. The experimental protocol for using deidentified human molar teeth was reviewed and approved by the Biomedical Institutional Review Board at Creighton University, Omaha, NE, USA (No. 760765-1). The enamel bonding sites were

Table 1: *Materials Used in This Study*

Materials (Lot No.)	Type of Material (Code)	Main Components	Manufacturer
Scotchbond Universal Adhesive (566724)	Universal adhesive (SU)	Bis-GMA, HEMA, MDP, ethyl methacryalate, methyl-reaction products with decanediol and phosphorous oxide, propenoic acid, copolymer of acrylic and itaconic acid, dimethylaminobenzoate, methyl ethyl ketone, ethanol, water, camphorquinone, silane-treated silica	3M ESPE, St Paul, MN, USA
Adhese Universal (164453)	Universal adhesive (AU)	BIS-GMA, HEMA, MDP, MCAP, decanediol dimethacrylate, dimethacrylate, ethanol, water, initiator, stabilizers, silicon dioxide	Ivoclar Vivadent, Schaan, Lichtenstein
G-Premio Bond (541424)	Universal adhesive (GB)	MDP, 4-MET, MEPS, methacrylate monomer, acetone, water, initiator, silica	GC, Tokyo, Japan
Ultra-Etch (G019)	Pre-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 ESPE, St Paul, MN, USA

Abbreviations: bis-GMA, bisphenol A glycidyl methacrylate; 4-MET, 4-methacryloyloxyethyl trimellitate; HEMA, 2-hydroxyethyl methacrylate; MCAP, methacrylated carboxylic acid polymer; MDP, 10-methacryloyloxydecyl di-hydrogen phosphate; MEPS, methacryloyloxyalkyl thiophosphate methylmethacrylate; TEGDMA, triethylene glycol dimethacrylate.

prepared by sectioning the teeth mesiodistally and removing approximately two-thirds of the apical root structure. Sectioned buccal and lingual halves were mounted in 25-mm brass rings with Bosworth Fastray acrylic material (Keystone Industries, Gibbstown, NJ, USA). The enamel surfaces were ground flat with 180-, 320-, 600-, 1200-, and 4000-grit silicon carbide paper (Struers, Cleveland, OH, USA) using a grinder-polisher (Ecomet 4, Buehler, Lake Bluff, IL, USA). These surfaces were then washed and dried using a dental three-way syringe at a distance of 5 cm above the surface at air pressure of 0.3 MPa. Some enamel surfaces were pre-etched with phosphoric acid for 15 seconds prior to application of the adhesive (with pre-etching), whereas others were not (without pre-etching). The specimens were prepared under ambient conditions of 23°C ± 2°C and 50% ± 10% relative humidity.

Fifteen specimens per group were used to determine the initial shear bond strength of the universal adhesive. The adhesives were applied to the enamel surfaces according to the manufacturers' instruc-

Table 2: *Application Protocol for Phosphoric Acid Pre-Etching, Universal Adhesives, and Protocol for Making Specimens of Presence- and Absence-of-Oxygen-Inhibition-Layer Groups*

Component	Protocol
Pre-etching	Pre-etching
With pre-etching	Enamel surface was acid etched with phosphoric acid for 15 s, rinsed with water for 15 s (three-way dental syringe) and air-dried
Without pre-etching	Phosphoric acid pre-etching was not performed
Adhesive	Adhesive application
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. Adhesive photo-cured for 10 s.
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. Adhesive 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. Adhesive photo-cured for 10 s.
Oxygen inhibition	Making specimens
Presence of oxygen inhibition layer	The adhesives were applied to the enamel surfaces according to the manufacturer instructions and photo-cured for 10 s.
Absence of oxygen inhibition layer	The top surface of the specimens of presence group was removed with ethanol-impregnated cotton pads.

Abbreviations: AU, Adhese Universal; GB, G-Premio Bond; SU, Scotchbond Universal Adhesive.

tions (Table 2) and photo-cured for 10 seconds at a standardized distance of 1 mm with a quartz-tungsten halogen unit (Spectrum 800 Curing Unit, Dentsply Caulk, Milford, DE, USA) set at 600 mW/cm^2 (presence-of-oxygen-inhibition-layer group). To produce specimens without an oxygen inhibition layer (absence-of-oxygen-inhibition-layer group), the top surface of the specimens was treated with ethanol-impregnated cotton pads (Medline Sterile Alcohol Prep Pads, Medline Industries, Mundelein, IL, USA).

Metal rings machined from 304 stainless steel and with an inner diameter of 2.4 mm, outer diameter of 4.8 mm, and height of 2.6 mm were used to confine the resin composite on the enamel surfaces for shear bond strength tests. Following the application of adhesive to the bonding sites, a releasing agent (3% solution of paraffin in hexane) was applied to the contact surface of the metal ring, which was positioned over the bonding site and secured in place by clamping in a custom fixture. The resin composite was filled in the ring and photo-cured for 30 seconds at a standardized distance of 2 mm, resulting in a resin composite cylinder measuring 2.4 mm in diameter and 2.5 mm in height inside the ring. The ring was left in place for the tests, and the bonded specimens were stored for 24 hours in distilled water at 37°C before testing.

The specimens were loaded to failure at 1.0 mm per minute using an ElectroPuls E1000 machine (Instron Worldwide Headquarters, Norwood, MA, USA). A metal rod with a chisel-shaped end was used to apply the load to the metal ring immediately adjacent to the flat-ground enamel surface. The initial shear bond strength (MPa) was calculated by dividing the peak load at failure by the bonded surface area. After testing, the bonding site tooth surfaces and resin composite cylinders were observed under an optical microscope (MZ16, Leica Microsystems, Buffalo Grove, IL, USA) at a magnification of 20× to determine the bond failure mode. The proportion of the resin composite surface with adherent enamel and visible residues was estimated to determine the type of failure (adhesive failure, cohesive failure in enamel, cohesive failure in resin composite, or mixed failure [combination of adhesive and cohesive failure]).

Shear Fatigue Strength Tests

The staircase method was used for shear fatigue strength tests,[22,23] and the specimens were prepared in the same way as they were for initial shear bond strength tests. The lower load limit was set near zero (0.4 N) and the setting between 50% and 60% of the initial shear bond strength determined for each of the adhesives tested was used for the initial maximum load. The load was applied at a frequency of 20 Hz using an electronic dynamic test instrument (ElectroPuls E1000, Instron Worldwide Headquarters) with a sine wave for 50,000 cycles or until failure occurred. The specimens were immersed in water at approximately 23°C to minimize the temperature rise, which might influence the mechanical properties. The load was incrementally (approximately 10% of the initial load) adjusted upward or downward (depending on survival or failure, respectively), and 20 specimens were used to determine the shear fatigue strength under each test condition. Using the calculation described by Draughn[22] and Dewji and others,[23] the test stress likely to produce 50% failure was termed *shear fatigue strength*. The mean shear fatigue strength (X) and its standard deviation (S) were calculated by the following formulae.

$$\mathrm{X} = \mathrm{Xo} + \mathrm{d}\left(\frac{A}{N} - \frac{1}{2}\right)$$

$$\mathrm{S} = 1.62\mathrm{d}\left(\frac{NB - A^2}{N^2} + 0.029\right)$$

$$\mathrm{N} = \sum n_i,\ \mathrm{A} = \sum i n_i,\ \mathrm{B} = \sum i^2 n_i,$$

where Xo is the lowest stress level considered in the analysis, and d is the stress increment used in the sequential tests. The lowest stress level at which a failure occurs is denoted by i = 0, the next i = 1, etc; n_{ii} is the number of failures after shear bond fatigue strength tests in each increment.

After testing, the type of bond failure was examined in the same manner.

Water Contact Angle Measurements

Specimens of the presence and absence groups with and without pre-etching were prepared as described for initial shear bond strength tests. The equilibrium water contact angles were measured in 10 specimens per group by the sessile drop method under ambient conditions of 23°C ± 2°C and 50% ± 10% relative humidity using a contact angle measurement apparatus (DM 500, Kyowa Interface Science, Saitama, Japan) fitted with a charge-coupled device camera to enable automatic measurement. A standardized 3 μL drop of distilled water was placed on the cured adhesive surface, and a profile image was captured after 500 milliseconds using the apparatus. Water

Table 3: *Initial Shear Bond Strengths (MPa) and Standard Deviations (in Parentheses) for Universal Adhesives*

Oxygen Inhibition	With Pre-Etching[a]			Without Pre-Etching		
	SU	AU	GB	SU	AU	GB
Presence of oxygen inhibition layer group	44.3 (4.8) a,A	40.6 (3.9) a,A	42.4 (5.5) a,A	27.2 (2.3) a,B	25.4 (3.5) a,B	26.1 (2.4) a,B
Absence of oxygen inhibition layer group	36.6 (4.2) b,A	34.8 (4.4) b,A	34.1 (3.1) b,A	22.0 (3.0) b,B	20.3 (3.7) b,B	20.4 (3.1) b,B

Abbreviations: AU, Adhese Universal; GB, G-Premio Bond; SU, Scotchbond Universal Adhesive.
[a] Same small letter in same column indicates no significant difference ($p>0.05$). Same capital letter within individual rows indicates no significant difference ($p>0.05$).

contact angles were then calculated by the θ/2 method using the built-in interface measurement and analysis system (FAMAS, Kyowa Interface Science).

Scanning Electron Microscopy Observation of Fracture Surface

Representative images of the fracture surfaces after initial shear bond strength and shear fatigue strength tests were taken in three specimens per group using scanning electron microscopy (SEM; TM3000 Tabletop Microscope, Hitachi High Technologies, Tokyo, Japan). The specimens were coated with a thin film of gold-palladium in a vacuum evaporator (Emitech SC7620 Mini Sputter Coater, Quorum Technologies, Ashford, UK), and SEM observations were carried out using an operating voltage of 15 kV.

SEM Observation of Resin-Enamel Interface

Representative images of the resin-enamel interfaces were carried out in three specimens per group using field-emission SEM (ERA 8800FE, Elionix, Tokyo, Japan). Bonded specimens of each group were stored in distilled water at 37°C for 24 hours, embedded in self-curing epoxy resin (Epon 812, Nisshin EM, Tokyo, Japan), and then stored at 37°C for a further 24 hours. They were then sectioned along the diameter of the resin composite post, and the surfaces of the cut halves were polished with 180-, 320-, 600-, 1200-, and 4000-grit silicon carbide paper using a grinder-polisher (Ecomet 4, Buehler). The surface was finally polished with a soft cloth using 1.0-μm-grit diamond paste. SEM specimens of the resin-enamel interface were dehydrated by first immersing them in ascending concentrations of aqueous tert-butanol (50% for 20 minutes, 75% for 20 minutes, 95% for 20 minutes, and 100% for two hours) and then transferring them to a critical-point dryer (Model ID-3, Elionix, Tokyo, Japan) for 30 minutes. To enhance the visibility of the layers, the polished surfaces were etched for 40 seconds using an argon ion beam (Type EIS-200ER, Elionix) directed perpendicularly to the surface at an accelerating voltage of 1.0 kV and an ion current density[24] of 0.4 mA/cm^2. Surfaces were coated with a thin film of gold in a vacuum evaporator (Quick Coater Type SC-701, Sanyu Electron, Tokyo, Japan). SEM observations were carried out using an operating voltage of 10 kV.

Statistical Analysis

All statistical tests were carried out using a commercial statistical software package (SPSS Statistics Base for Windows, IBM, Armonk, NY, USA), except for the modified *t*-test with Bonferroni correction, which was performed using custom software. Because the shear bond strength and water contact angle data exhibited normal distribution (Kolmogorov-Smirnov test), a three-way analysis of variance (ANOVA) and Tukey post hoc test at a significance level of 0.05 were used to analyze differences.

RESULTS

Initial Shear Bond Strength Tests

The initial shear bond strengths of universal adhesives in the presence and absence groups with and without pre-etching are listed in Table 3. The three-way ANOVA revealed that the presence or absence of an oxygen inhibition layer, adhesive type, and etching mode had a significant influence ($p<0.05$) on shear bond strength, but the interactions among these factors were not significant ($p>0.05$).

Regardless of etching mode, the initial shear bond strengths in the presence-of-oxygen-inhibition-layer group were significantly higher ($p<0.05$) than those in the absence group. Moreover, universal adhesives with pre-etching exhibited significantly higher ($p<0.05$) initial shear bond strengths than those without pre-etching, regardless of the presence or absence of oxygen inhibition layer and adhesive type.

Shear Fatigue Strength Tests

Shear fatigue strengths of universal adhesives in the presence and absence groups with and without pre-

Table 4: *Shear Fatigue Strengths (MPa) and Standard Deviations (in Parentheses) for Universal Adhesives*

Group	With Pre-Etching [a]			Without Pre-Etching		
	SU	AU	GB	SU	AU	GB
Presence of oxygen inhibition layer	22.2 (2.6) a,A	20.3 (2.3) a,A	21.1 (2.1) a,A	13.8 (2.7) a,B	12.6 (1.6) a,B	13.8 (2.4) a,B
Absence of oxygen inhibition layer	18.1 (2.2) b,A	17.1 (2.5) b,A	16.0 (2.1) b,A	11.0 (1.0) a,B	9.8 (0.8) b,B	10.3 (1.0) b,B

Abbreviations: AU, Adhese Universal; GB, G-Premio Bond; SU, Scotchbond Universal Adhesive.
[a] Same capital letter within individual rows indicates no significant difference ($p>0.05$).

etching are listed in Table 4. Regardless of the etching mode, the shear fatigue strengths of the presence-of-oxygen-inhibition-layer group were significantly higher ($p<0.05$) than those of the absence group. Moreover, universal adhesives with pre-etching exhibited significantly higher ($p<0.05$) shear fatigue strengths than those without pre-etching, regardless of the presence or absence of an oxygen inhibition layer and adhesive type.

Fracture Mode Analysis of Debonded Specimens

The failure modes of debonded specimens after initial shear bond strength and shear bond strength tests are shown in Table 5. The predominant mode of failure observed was the adhesive type, and a χ^2 test showed no significant differences in failure mode with presence or absence of an oxygen inhibition layer, adhesive type, or etching mode.

Water Contact Angle Measurement

The water contact angles of universal adhesives in the presence and absence groups with and without pre-etching are listed in Table 6. The three-way ANOVA test revealed that the presence or absence of an oxygen inhibition layer and adhesive type exerted a significant influence ($p<0.05$) on water contact angles, whereas etching mode had no such influence ($p>0.05$). The interaction between the presence or absence of an oxygen inhibition layer and adhesive type was also more significant than other interactions for water contact angles.

The water contact angles of the presence-of-oxygen-inhibition-layer group were significantly lower ($p<0.05$) than those of the absence group, regardless of the adhesive type and etching mode. Moreover, the water contact angles of universal adhesives in the presence and absence groups were also material dependent.

SEM Observation of Fracture Surface

Representative SEM images of the debonded specimens after shear fatigue strength tests are shown in Figure 1 (images after initial shear bond strength tests were similar). The specimens predominantly exhibited adhesive failure at a lower magnification, and the specimens with pre-etching showed more enamel fragments than those without pre-etching at higher magnification, regardless of the presence or absence of an oxygen inhibition layer.

SEM of Resin-Enamel Interface

Representative SEM images of the resin-enamel interfaces are shown in Figure 2. The resin-enamel interfaces of all the adhesives showed excellent adaptation, regardless of which group they belonged to. Moreover, the thickness of the adhesive layer in the presence-of-oxygen-inhibition-layer group ap-

Table 5: *Failure Mode Analysis of Debonded Specimens After Initial Shear Bond Strength and Shear Fatigue Strength Tests[a]*

Bond Strength Test	Group	Pre-Etching	SU	AU	GB
Initial shear bond strength	Presence of oxygen inhibition layer	With pre-etching	[67/0/13/20]	[93/0/7/0]	[86/0/7/7]
		Without pre-etching	[100/0/0/0]	[100/0/0/0]	[100/0/0/0]
	Absence of oxygen inhibition layer	With pre-etching	[86/7/7/0]	[100/0/0/0]	[100/0/0/0]
		Without pre-etching	[100/0/0/0]	[100/0/0/0]	[100/0/0/0]
Shear fatigue strength	Presence of oxygen inhibition layer group	With pre-etching	[60/10/20/10]	[90/0/10/0]	[80/0/10/10]
		Without pre-etching	[100/0/0/0]	[100/0/0/0]	[90/0/10/0]
	Absence of oxygen inhibition layer group	With pre-etching	[80/0/15/5]	[90/0/10/0]	[85/0/5/10]
		Without pre-etching	[100/0/0/0]	[100/0/0/0]	[100/0/0/0]

Abbreviations: AU, Adhese Universal; GB, G-Premio Bond; SU, Scotchbond Universal Adhesive.
[a] Percentage of failure mode [adhesive failure/cohesive failure in resin/cohesive failure in enamel/mixed failure]. There was no significant differences in failure mode with presence or absence of oxygen inhibition layer, type of adhesive, or pre-etching status.

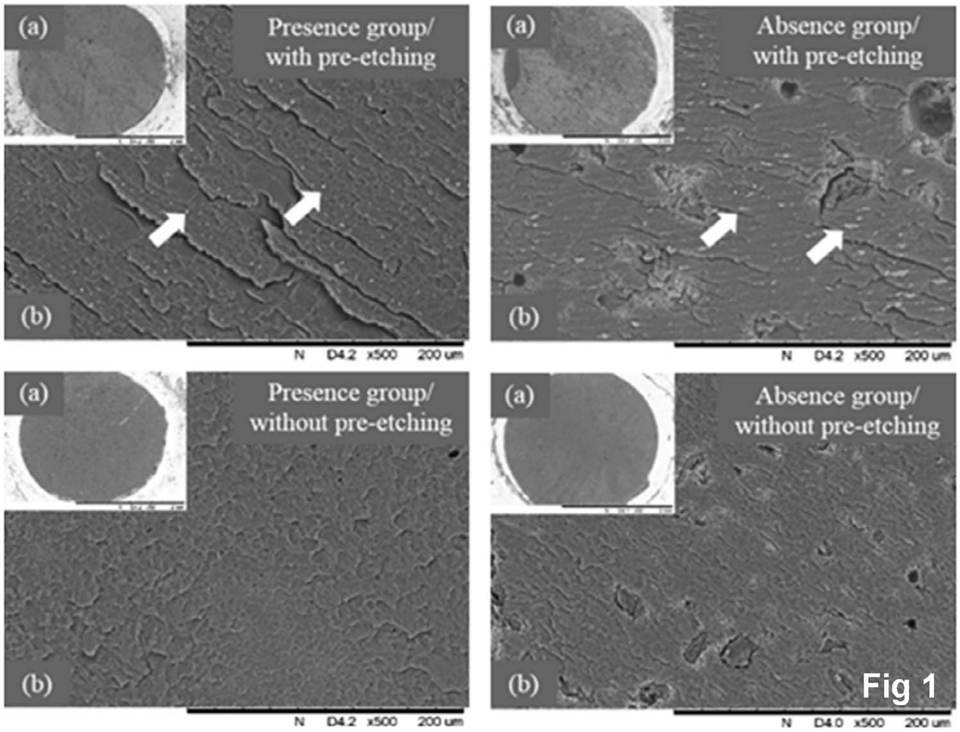

Figure 1. *Representative scanning electron microscopy images of debonded specimens after shear fatigue strength tests at (a): 50× magnification and (b): 500× magnification. The debonded specimens predominantly exhibited adhesive failure at lower magnification, and cracks and cleavages could be clearly seen in the specimens at higher magnification. Some features of enamel fragments such as benchmarks (arrows) can be observed in the specimens with phosphoric acid pre-etching, indicating strong bonding between the enamel and adhesive.*

proximated 9 μm, whereas that of the absence group was approximately 4 μm, irrespective of whether they had undergone pre-etching.

DISCUSSION

The present study indicates that the initial shear bond strengths and shear fatigue strengths of universal adhesives in the presence-of-oxygen-inhibition-layer group were significantly higher than those in the absence group, regardless of adhesive type and etching mode. Bond fatigue strength testing is one way to investigate clinical bond fatigue durability for *in vitro* studies, given that bonded specimens are subjected to repeated subcritical loading challenges, simulating intraoral conditions.[25,26] Therefore, the results of the current study indicate that the oxygen inhibition layer of universal adhesives may not impair enamel bonding in terms of bond fatigue durability. However, there were no significant differences in failure type between the presence and absence groups, and adhesive failure was predominant in both initial shear bond strength and shear fatigue strength tests. In the present study, the shear load was applied to the metal rings enclosing the resin composite bonded to enamel, and adhesive failures were observed most frequently, regardless of the presence or absence of an oxygen inhibition layer. This can be explained by the fact that bonding specimens enclosed within a metal mold exhibit a higher frequency of adhesive failure than unenclosed specimens.[27,28] Scherrer and others[29] argued that an increased production of adhesive failure in bond strength tests was desirable to provide more relevant information about the mean bond strength. Therefore, in this study, the type of fracture observed may provide evidence on the relevance of the initial shear bond strengths and shear fatigue strengths of universal adhesives.

Table 6: *Water Contact Angles (°) of the Universal Adhesives Under All Experimental Conditions[a]*

Group	With Pre-Etching			Without Pre-Etching		
	SU	AU	GB	SU	AU	GB
Presence of oxygen inhibition layer	51.1 (3.8) a,A	49.2 (3.1) a,A,B	46.4 (3.9) a,B	51.4 (3.2) a,A	49.1 (3.5) a,A,B	47.9 (3.0) a,B
Absence of oxygen inhibition layer	58.5 (2.4) b,A	55.1 (2.1) b,A,B	54.3 (2.6) b,B	57.9 (2.5) b,A	55.8 (3.0) b,A,B	53.7 (2.2) b,B

Abbreviations: AU, Adhese Universal; GB, G-Premio Bond; SU, Scotchbond Universal Adhesive.
[a] *Values in parentheses are standard deviations. Same small letter in same column indicates no significant difference ($p > 0.05$). Same capital letter within individual rows indicates no significant difference ($p > 0.05$).*

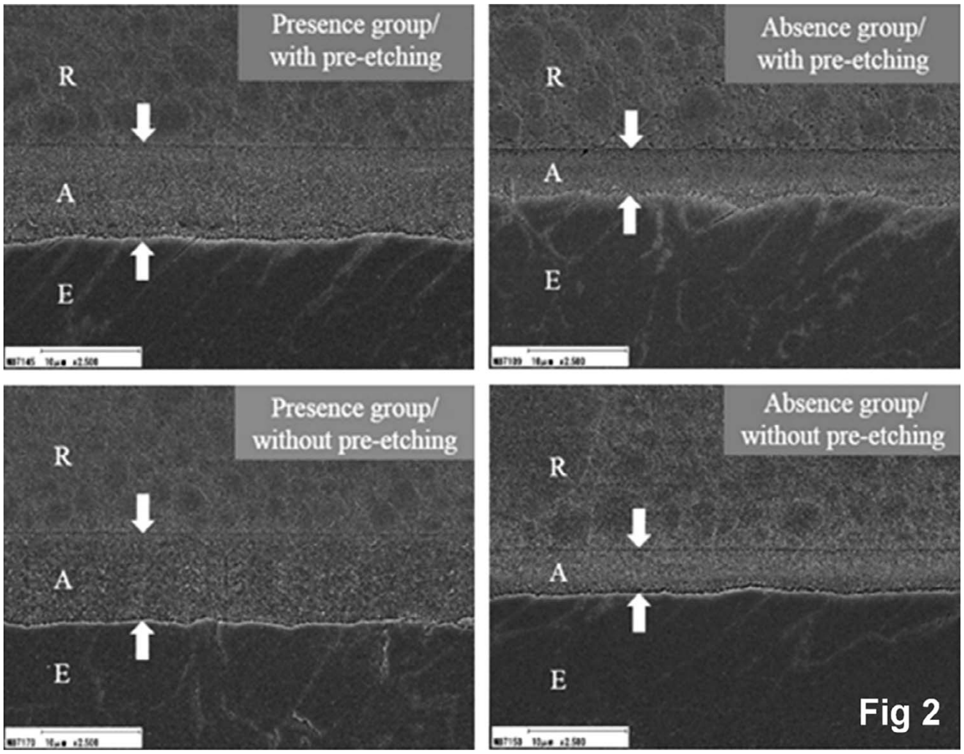

Figure 2. *Representative field-emission electron microscopy images of the resin-enamel interface at 2500× magnification. The resin-enamel interface of the universal adhesives used showed excellent adaptation, regardless of the presence or absence of an oxygen inhibition layer. Arrows indicate the thickness of the adhesive layer. Regardless of the etching mode, the thickness of the adhesive layer of the specimens in the presence-of-oxygen-inhibition-layer-group was approximately 9 µm, whereas that of the absence group was approximately 4 µm. Abbreviations: A, adhesive; E, enamel; R, resin composite.*

Interfacial characteristics in terms of water contact angle were also investigated to understand their relationship with the results of the initial shear bond strength and shear fatigue strength tests. Cured universal adhesives in the presence-of-oxygen-inhibition-layer group exhibited significantly lower water contact angles than those in the absence group, demonstrating their greater polarity and hydrophilicity. During the polymerization reaction, monomers in universal adhesives form highly cross-linked polymeric networks, which diminish their polarity and hydrophilicity.[30] Therefore, if the polymerization reaction of monomers in universal adhesives progresses to completion, the polarity and hydrophilicity should be reduced in the cured adhesive. Conversely, incomplete polymerization may lead to residual functional monomers within the adhesive.[31] Therefore, oxygen impairment of cross-linking within the oxygen inhibition layer of a cured adhesive may have led to lower water contact angles in the presence group.

In addition, the water contact angles of universal adhesives increased when the oxygen inhibition layer was removed, whereas the initial shear bond strengths and shear fatigue strengths decreased. Optimal wettability is important to enable the materials to spread across the entire surface and establish adhesion.[32] Thus, the maximum bond strength is assumed to occur when the wettability of the adherent surface is maximized.[33] This suggests that the interfacial characteristics of the universal adhesives strongly influenced the results of the initial shear bond strength and shear fatigue strength tests. Moreover, the oxygen inhibition layer readily adapts to the overlying material to increase the contact area; it also allows the materials on both sides to cross the interface and blend together to form an interdiffused zone where copolymerization can take place to produce a chemical bond between residual functional monomers in the oxygen-inhibition layer and the overlying resin composite.[7] All these effects tend to strengthen the layer-layer interaction.[34] This may explain why the cured universal adhesive in the presence-of-oxygen-inhibition-layer group with lower water contact angles exhibited higher initial shear bond strengths and shear fatigue strengths. Furthermore, the thickness of the universal adhesive layer of the presence group was approximately 9 µm, whereas that of the absence group was 4 µm, as seen in SEM observation of the resin-enamel interfaces. The relatively thin oxygen inhibition layer may have allowed complete diffusion of the photo-initiator into the overlying composite, resulting in higher initial shear bond strengths and shear fatigue strengths.

On the basis of the results of this study, our null hypothesis (that the enamel bond fatigue durability and interfacial characteristics of universal adhesives would not be influenced by the presence or absence of the oxygen inhibition layer) was rejected.

CONCLUSION

The results of this study indicate that the oxygen inhibition layer of universal adhesives significantly increases the enamel bond fatigue durability and greatly changes interfacial characteristics, suggesting that the bond fatigue durability and interfacial characteristics of these adhesives strongly rely on its presence.

Acknowledgement

The authors thank Mr Jason M Moody for technical contributions.

Regulatory Statement

This study was conducted in accordance with all the provisions of the local human subjects oversight committee guidelines and policies of the Biomedical Institutional Review Board at Creighton University, Omaha, NE, USA. The approval code for this study is 760765-1.

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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