Compatibility of new universal simplified adhesives with dual-cured composite and effect of hydrophobic resin coat

Kun-Suk Jeong

Department of Dental Science

The Graduate School, Yonsei University

Compatibility of new universal simplified adhesives with dual-cured composite and effect of hydrophobic resin coat

Directed by Professor Byoung-Duck Roh

The Master's Thesis
submitted to the Department of Dentistry,
the Graduate School of Yonsei University
in partial fulfillment of the requirements for the degree of
Master of Dental Science

Kun-Suk Jeong

June 2014

This certifies that the Master's Thesis of
Kun-Suk Jeong is approved.

Byoung-Duck Roh

Sung-Ho Park

Jeong-Won Park

The Graduate School
Yonsei University

June 2014

감사의 글

보존과에서 새 출발할 때의 설렘이 아직 생생한데 어느새 3년차가 되어 학위논문이라는 큰 결실을 맺게 되었습니다. 아직 부족하고 서툴지만 이렇게 뜻깊은 결실을 맺는데 도움과 의지가 되어 주신 많은 분들께 감사의 말씀을 전하고자 합니다.

먼저, 든든한 버팀목이 되어 주시고 막막했던 논문의 방향을 제시해주시며, 따뜻한 격려를 아끼지 않으셨던 지도교수 노병덕 교수님께 진심으로 감사 드립니다. 항상 애정과 관심을 보이시며 실험을 진행하는데 많은 도움을 주신 박성호 교수님과 완성도 높은 논문을 위해 꼼꼼한 수정과 관심을 아끼지 않으신 박정원 교수님께도 깊은 감사의 말씀을 드립니다. 또한 보존과의 멘토이신 이찬영 교수님, 이승종 교수님, 수련 교육부터 인생의 조언까지 자식처럼 챙겨주신 김의성 교수님, 정일영 교수님, 신유석 교수님께도 감사 드립니다. 저를 훌륭한 보존과 치과의사로 수련시켜 주신 보존과 모든 교수님께 정말 감사드리며, 늘 겸손하고 꾸준히 노력하는 모습 보이도록 하겠습니다.

그 동안 함께 생활하며 즐거운 추억 만들어준 1,2년차 선생님들에게도 고마운 마음을 전합니다. 무엇보다 오랜 시간을 함께 기쁨과 슬픔을 나누며 의지가 되어준 수련동기들. 지금의 동기들이 있었기에 제가 이 자리에 있다고 생각하며, 그 동안 말 못한 고마움을 표현합니다.

마지막으로 객지 생활하는 아들을 걱정하고 뒷바라지하느라 늘 희생하시는 사랑하는 부모님과 부족한 형을 자랑스럽게 여기고 응원해주는 든든한 동생 건욱이에게 표현하지 못한 사랑하는 마음을 전합니다.

2014년 6월

정 건 석

Table of Contents

List of Figures

List of Tables

Abstract

Compatibility of new universal simplified adhesives with dual-cured composite and effect of hydrophobic resin coat

Kun-Suk Jeong, D.D.S.

Department of Dental Science, The Graduate School, Yonsei University

(Directed by Prof. Byoung-Duck Roh, D.D.S., M.S., Ph.D.)

1. Objective

The purpose of this study was to evaluate the influence of two different universal adhesive on microshear bond strength of dual-cured composite to dentin. Also we evaluated the effect of application of a hydrophobic resin coat on the bond strength within the hybrid layer of a one-step self-etch adhesive system.

2. Materials and methods

The flat dentin surfaces were prepared from freshly extracted, caries-free, human third molars. Four recent adhesive systems including two universal adhesive (Single Bond Universal, All-Bond Universal), one-step self-etch system (Tetric N-Bond Self-Etch), etch-and-rinse system (XP bond) and hydrophobic adhesive resin for coating

(Heliobond) were evaluated with dual-cured type composite. As a control group, light-cured type flowable composite, with similar flexural strength as dual-cured composite was selected.

Microshear bond test was carried out using a universal testing machine and all debonded specimens with dual-cured composite were observed under X40 stereoscope to determine the mode of failure. Furthermore, specimens in each group with different adhesives were observed under scanning electron microscope for detailed evaluation of surface morphology.

3. Result

Single Bond Universal exhibited the highest bond strength (28.5 ± 6.2 MPa). On the other hand, All-Bond Universal showed low microshear bond strength (14.1 ± 5.1 MPa) and statistically no difference with Tetric N-Bond Self-Etch (8.8 ± 6.7 MPa). Also, with an additional hydrophobic resin coat, Single Bond Universal exhibited significantly higher bond strength than All-Bond Universal. When used with an additional hydrophobic resin coat, the bond strength of all adhesives increased significantly except for Single Bond Universal.

The failure mode of Single Bond Universal was predominantly mixed failure. In case of other adhesives, high percentages of adhesive failure appeared. When a hydrophobic resin coat was applied, Single Bond Universal and All-Bond Universal did not show much difference, whereas Tetric N-bond Self-etch and XP bond showed an increased percentage of mixed failure.

4. Conclusion

(1) With respect to the influence of universal adhesives, Single Bond Universal showed the highest microshear bond strength but not incompatibility with dual-cured composite. However All-Bond Universal showed relatively low microshear bond strength and statistically no difference with Tetric N-Bond Self-Etch as existing one-step self-etch system.

(2) Hydrophobic resin coat may be considerable alternation for solving the problem of incompatibility between single step adhesive system and dual-cured composite, and can improve microshear bond strength of these materials.

Key words: microshear bond strength, universal adhesive, self-etch adhesive, dual-cured composite, compatibility, incompatibility, hydrophobic resin coat

Compatibility of new universal simplified adhesives with dual-cured composite and effect of hydrophobic resin coat

Kun-Suk Jeong

Department of Dental Science

The Graduate School, Yonsei University

(Directed by Prof. Byoung-Duck Roh)

I. Introduction

Contemporary dentin adhesives are classified as three-step, two-step and one-step systems, depending on how the three principle steps of etching, priming, and bonding to tooth substrates are accomplished or simplified (Perdigao, 2007). Recently, various adhesive systems have been developed for simplification and reduced application time.

Dentin adhesive is used for a great variety in clinical applications ranging from direct restorations, core build-up restorations to the adhesive cementation of indirect restorations and fiber-reinforced root canal post. Thus, it is clear that the sometimes confusing variety of different types of bonding systems has to be considered when assessing their compatibility for clinical use (Haller, 2013).

Swift et al. noted that there is incompatibility between simplified-step adhesives and chemically or dual-cured composites (Swift et al., 1998). Also, Moll K. reported that all-in-one adhesives are not suitable for the combination with chemically cured or dual-cured composites, as a result, the bond strength to dentin is dramatically reduced (Moll et al., 2007). Studies have suggested that adverse interactions occur between dual-cured or chemically cured resin composites and uncured acidic resin monomers in the underlying self-etching adhesive layer (Giannini et al., 2004; Swift et al., 1998). This uncured monomer surface layer is formed due to atmospheric oxygen inhibition effects on the activation of the adhesive resin. The adverse chemical interaction between unpolymerized acidic resin monomers in the adhesive and the basic tertiary amine catalyst in the composite was thought to be responsible for the observed incompatibility (Schittly et al., 2010). Sanares et al. explained that the interaction between acidic adhesive resin monomers and the tertiary amines of the composites results in consumption of the latter in acid-base reactions, depriving them of their capacity to generate free radical in subsequent redox reactions (Sanares et al., 2001).

A different mechanism was proposed to explain the adverse interaction. It has been shown that single-step, self-etching adhesives permit the passage of fluid and behave as semipermeable membranes after polymerization (Chen and Suh, 2013; Tay et al., 2003a). These adhesives have hydrophilic nature and exhibit complicated patterns of water-filled channels within the adhesive layer. The water moves to the composite-adhesive interface, resulting in mechanical disruption of the coupling between the adhesive and composite resin

In order to overcome these adverse interaction, Tay et al. suggested that the use of a chemical co-initiator could solve this incompatibility problem between self-etching adhesive and chemically cured composite, which presumably replenishes free radicals that were depleted from the tertiary amine of the chemically cured composite by reacting with residual acidic resin monomers (Tay et al., 2003a). Some alternative initiators such as sulphinic acid salts, organoboron compounds, and barbituric acid/cupric chloride have been included in the adhesives (Imai et al., 1991; Nyunt and Imai, 1996). Many manufacturers provide a dual cure activator which contains sulphinic acid salts by mixing with the simplified acidic adhesives to improve their incompatibility with dual/self-cured composite. Other manufacturers create amine-free dual cure product (eg. NX3, Kerr) to avoid theirs incompatibility. Also, other studies proposed that the use of either an additional adhesive resin layer without acidic monomers or an intermediate low viscosity composite liner could enhance bond strength between the adhesive and resin composite (King et al., 2005; Perdigao et al., 2013). Giannini et al. showed that the use of an additional composite liner could solve this incompatibility, significantly improving bond strength between self-etch adhesive and dual-cured composite (Giannini et al., 2004). Thus, the application of a hydrophobic resin coat is expected to be a clinically favorable method to overcome this incompatibility.

Recently, some manufacturers have introduced more versatile adhesive system that are called “universal”, “multi-purpose” or “multi-mode” adhesive and can be applied as self-etch or as etch-and-rinse adhesive. These novel universal multimode adhesives have been developed to make the clinical procedure more simple and user-friendly. Furthermore, these new adhesives contain 10-methacryloyloxydecyl dihydrogen phosphate (MDP) monomers, which bond chemically to dentin and enhance durability (Inoue et al., 2005). The advantage of using a universal adhesive is suitable for a great range of restoration materials. Some manufacturers propose that the ultra-mild acidity (pH > 3) allows the universal adhesive to be compatible with dual-cured and self-cured materials.

However, there is little information in the literature about the compatibility of these new universal adhesive to dual-cured composite. Thus, the purpose of this article was to evaluate the influence of two different universal adhesive on microshear bond strength of dual-cured composite to dentin. Also we evaluated the effect of application of a hydrophobic resin coat on the bond strength within the hybrid layer of a one-step self-etch adhesive system. The null hypotheses tested were that 1) there was no different on microshear bond strength of dual-cured composite to dentin with different bonding systems including universal adhesive; 2) the application of a hydrophobic resin coat would not influence the microshear bond strength of the adhesive systems to dentin.

II. Materials and Methods

1. Materials

Four recent adhesive systems including universal adhesive and dual-cured type composite resin were prepared. As a control group, light-cured type flowable composite, with similar flexural strength as dual-cured composite was selected. Two universal adhesives (Single Bond Universal, All-Bond Universal), one-step self-etch system (Tetric N-Bond Self-Etch), etch-and-rinse system (XP bond) and hydrophobic adhesive resin for coating (Heliobond) were evaluated. The materials employed in this study are listed in Table 1 and 2. These materials were applied according to the manufacturer's instructions.

Table 1. Composite resin

Product	Composition	Shade	Lot	Type	Manufacturer
LuxaCore® Dual	Barium glass 69%, pyrog. silica 3% in a Bis-GMA	A3	700585	Dual-cured	DMG
Metafil Flo	2,2-Bis[4-(methacryloxy-polyethoxy)phenyl-]propane, Other bi-functional methacrylate monomers, Barium/silica-glass, Silica, amorphous, 3-(trimethoxysilyl) propyl methacrylate	A3	FX14	Light-cured	SUN MEDICAL

Table 2. Adhesive Resin

Product	Composition	Lot	Manufacturer	Instruction for use
Single Bond Universal	MDP Phosphate Monomer, Dimethacrylate resins, HEMA, Vitrebond™ Copolymer, Filler, Ethanol, Water, Initiators, silane	517709	3M EPSE	1. Apply the adhesive to the entire preparation with a microbrush and rub it in for 20 s. If necessary, rewet the disposable applicator during treatment 2. Direct a gentle stream of air over the liquid for about 5 s until it no longer moves and the solvent has evaporated completely
All-Bond Universal	MDP, Ethanol, Bis-phenol glycidyl methacrylate (Bis-GMA), HEMA, water, initiators	1300000367	Bisco	1. Apply two separate coats of adhesive, scrubbing the preparation with a microbrush for 10-15 s per coat. Do not light polymerize between coats 2. Evaporate excess solvent by thoroughly air-drying with an air syringe for at least 10 s, there should be no visible movement of the material. The surface should have a uniform glossy appearance
Tetric N-Bond Self-Etch	derivatives of bisacrylamide, water, bis-methacrylamide dihydrogen phosphate, amino acid acrylamide, hydroxy alkyl methacrylamide, highly dispersed silicon dioxide, catalysts and stabilizers	R59913	Ivoclar Vivadent	1. Apply a thick layer of adhesive on the dentin surfaces and brush in for At least 30 s. All surfaces should be thoroughly coated. The total reaction time should be no shorter than 30 s. 2. Disperse excess amounts of adhesive with a strong stream of air until there is no longer any movement of the material
XP Bond	Carboxylic acid modified dimethacrylate (TCB resin); Phosphoric acid modified acrylate resin (PENTA); Urethane Dimethacrylate (UDMA); Triethyleneglycol dimethacrylate	1302000316	Dentsply	1. Dispense adhesive directly onto a fresh Applicator Tip or onto a disposable brush 2. Wet all cavity surfaces uniformly with adhesive, Avoid pooling 3. Leave the surface

	(TEGDMA); 2-hydroxyethylmethacrylate (HEMA); Butylated benzenediol (stabilizer); Ethyl-4-dimethylaminobenzoate; Camphorquinone; Functionalized amorphous silica; t-butanol			undisturbed for 20 s. 4. Evaporate solvent by thoroughly blowing with air from an air syringe for at least 5 s. The cavity surface should have a uniform, glossy appearance
Heliobond (for hydrophobic resin coat)	Bis-phenol glycidyl methacrylate(bis-GMA), triethylenedimethacrylate(TEGDMA)	P00765	Ivoclar Vivadent	Apply a thin layer of Heliobond onto the surface using a brush or spherical instrument. An optimal, thin layer can be achieved using a stream of air

2. Tooth specimen preparation

Tooth specimens were prepared from freshly extracted, caries-free, human third molars which were collected without any personal information. The teeth were disinfected and stored in distilled water. Horizontal dissections of the teeth were made at the cement-enamel junction and 3mm above this junction using a low speed precision diamond saw (TOPMET Metsaw-LS, R&B, Daejeon, Korea) under water cooling to obtain the specimens. The 3mm-thick dentin specimens were wet abraded with 600-grit SiC paper to create a standardized smear layer, and all the specimens were thoroughly rinsed with water.

3. Experimental groups

The prepared specimens were randomly divided into 12 groups, including a control group for using light-cured composite. The experimental groups tested in the study are summarized in Table 3. In each groups, 15 samples were assigned.

Table 3. Experimental groups **(N = 15)**

Adhesive system	Bond Strategy	Composite type	Hydrophobic resin coat
Single Bond Universal	Self-etch	Dual-cured	Without
			With
		Light-cured	Without(control)
All-Bond Universal	Self-etch	Dual-cured	Without
			With
		Light-cured	Without(control)
Tetric N-Bond Self-Etch	Self-etch	Dual-cured	Without
			With
		Light-cured	Without(control)
XP Bond	Etch & Rinse	Dual-cured	Without
			With
		Light-cured	Without(control)

4. Bonding procedure

The methodology used by Roh and Chung was performed to prepare specimens for the microshear bond test (Fig. 1) (Roh and Chung, 2005). Adhesives and composites were applied according to the manufacturer’s instructions.

In order to control the bonding area, acid/solvent-resistant adhesive tape (Scotch tape, 3M) with 4 holes (1.2mm diameter) was first attached to the dentin plates, completely covering the surface except for the hole area. Adhesives were applied over the tape and light cured for 20 seconds (600 mW/cm^2, Smart LED plus, Sungbotech, Seoul, Korea). In hydrophobic resin coat groups, an additional resin bonding agent (Heliobond, Ivoclar Vivadent) was applied after polymerization of previous adhesive and light cured for 20 seconds.

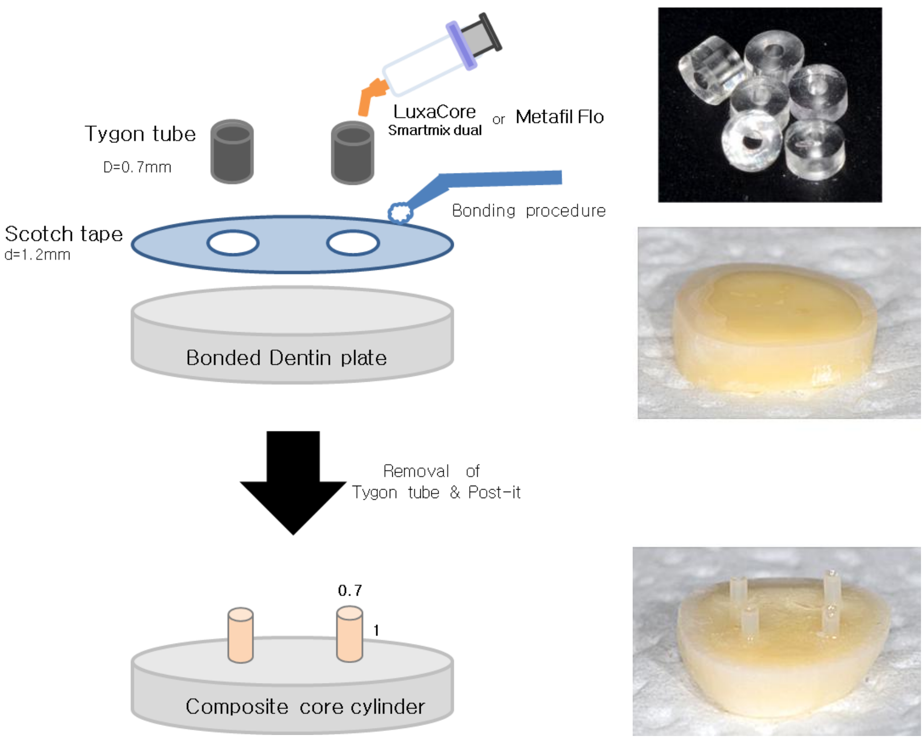


Fig. 1. Specimen Preparation

Cylindrical translucent molds (TYGON® R-3603 Laboratory Tubing, Saint Gobain performance Plastic, Maime Lakes, FL, USA) were prepared to control resin placement onto the dentin plate. The molds were positioned over the tape, ensuring that their lumen coincided with the circular bonded areas (Fig. 1). Dual-cured type core build up composite (LuxaCore® Dual, DMG, Germany) or light-cured type flowable composite (Metafil Flo, SUN MEDICAL, Japan) was applied into the molds to fill their internal volume, and immediately light cured for 20 seconds. The molds and tape were carefully removed, exposing the resin composite cylinders (0.7mm diameter, 1.0mm height) bonded to the dentin surface. Bonded area was meticulously checked under an optical microscope (X25) and specimens with gap, defect or void were excluded. The specimens were stored in distilled water at 37℃ for 24 hours

5. Microshear bond test

Following storage in distilled water at 37℃ for 24 hours, each specimen was attached to the testing device with cyanoacrylate glue (Zapit, DVA, Corona, CA, USA) and each composite core cylinder was tested. Microshear bond test was carried out using a universal testing machine (EZ-test 500, Shimazu, Tokyo, Japan) with cross-head speed of 0.5mm/min (Fig. 2). Shear load was applied to the base of the composite core cylinder with a thin metal wire (wire-loop method) until bond failure of the specimen occurred (Pashley et al., 1995). The load at failure was recorded in Newton (N) and converted to shear bond strength in MPa ($\tau=4P/\pi d^2$).

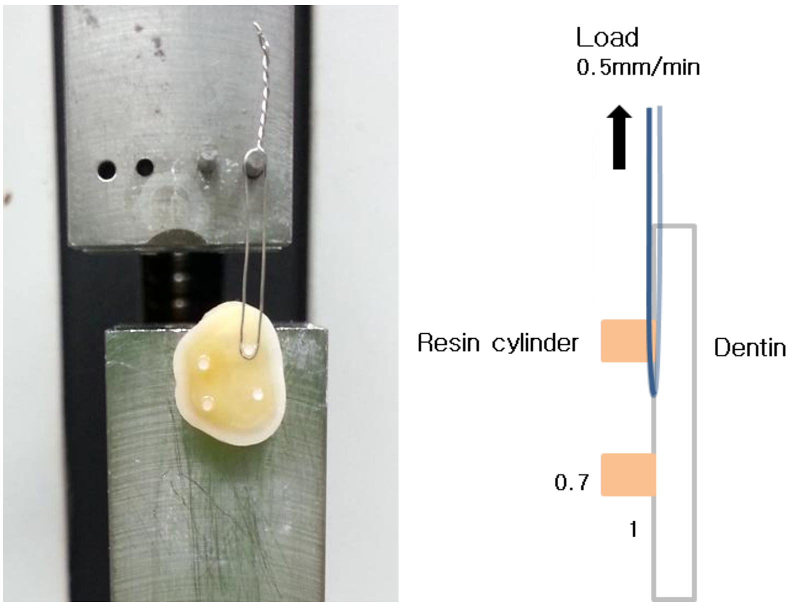


Fig. 2. Microshear bond test, wire-loop method.

6. Surface evaluation

All debonded specimens after shear bond test were observed under X40 stereoscope (Leica, Microsystems Inc., Depew, New York, USA) to determine the mode of failure. Failure modes were classified as follows:

- Adhesive failure at dentin-adhesive or adhesive-composite interface
- Cohesive failure within dentin or composite
- Mixed failure, involving bonding agent, composite and dentin interfaces.

Furthermore, specimens in each group with different adhesives were observed under scanning electron microscope (Hitachi, Tokyo, Japan) for detailed evaluation of surface morphology.

7. Statistical analysis

Statistical analyses were performed using SPSS 11.5 software for Windows (SPSS Inc., Chicago, IL, USA). One-way analysis of variance (ANOVA) was applied using microshear bond strength (MPa). Tukey's test was used in the post hoc comparisons. Statistical differences between the groups with and without a hydrophobic resin coat were analyzed using independent T-test. Including the control group, the microshear bond strength (MPa) of different composites using the same adhesive was reevaluated with one-way ANOVA. In all analyses, the level of significance was set at $\alpha = 0.05$.

III. Result

1. Microshear bond strength

The mean microshear bond strength and standard deviation values are summarized in Table 4. One-way ANOVA analysis indicated there was statistically significant difference among the mean bond strength of the adhesives ($F = 24.32$; $p < 0.0001$). Tukey's test showed that Single Bond Universal exhibited the highest bond strength ($p < 0.0001$). Also, with an additional hydrophobic resin coat, one-way ANOVA showed significant difference among the mean bond strength of the adhesive ($F = 5.43$; $p = 0.002$). According to Tukey's test, there was only a statistically significant difference between Single Bond Universal and All-Bond Universal, and Single Bond Universal exhibited higher bond strength than All-Bond Universal ($p < 0.001$). When used with an additional hydrophobic resin coat, independent T-test showed that the bond strength of all adhesives increased significantly except for Single Bond Universal.

In the control group, Single Bond Universal showed the highest bond strength, but there was no statistically significant difference in the other groups. When comparing the control group with the dual-cured composite group in each adhesive via one-way ANOVA, there was statistically significant difference between the groups. When verifying the results with Tukey's test, there was no notable difference between light-cured composite and dual-cured composite in the Single Bond Universal and XP bond group. On the other hand, in the All-Bond Universal group there was no notable difference between applying a hydrophobic resin coat and using light-cured composite; however, they showed higher bond strengths than using dual-cured composite only. The same applied to the Tetric N-Bond Self-Etch group.

Table 4. Microshear bond strength of adhesives (MPa ± SD) (N = 15)

Adhesive system	Mean bond strength (MPa)		
	Dual-cured composite		Light-cured composite
	Without hydrophobic resin coat	**With hydrophobic resin coat**	**Control**
Single Bond Universal	28.5 ± 6.2 A a	29.5 ± 6.4 A a	27.6 ± 5.2 A a
All-Bond Universal	14.1 ± 5.1 BC a	19.9 ± 5.5 B b	20.9 ± 5.4 B b
Tetric N-Bond Self-Etch	8.8 ± 6.7 C a	25.7 ± 6.0 AB b	18.6 ± 3.4 B c
XP Bond	16.7 ± 7.1 B a	24.4 ± 7.3 AB b	22.0 ± 4.4 B ab

Means followed by different letters (capital letter – column; lower case – line) differ significantly according to Tukey's test ($p < 0.05$).

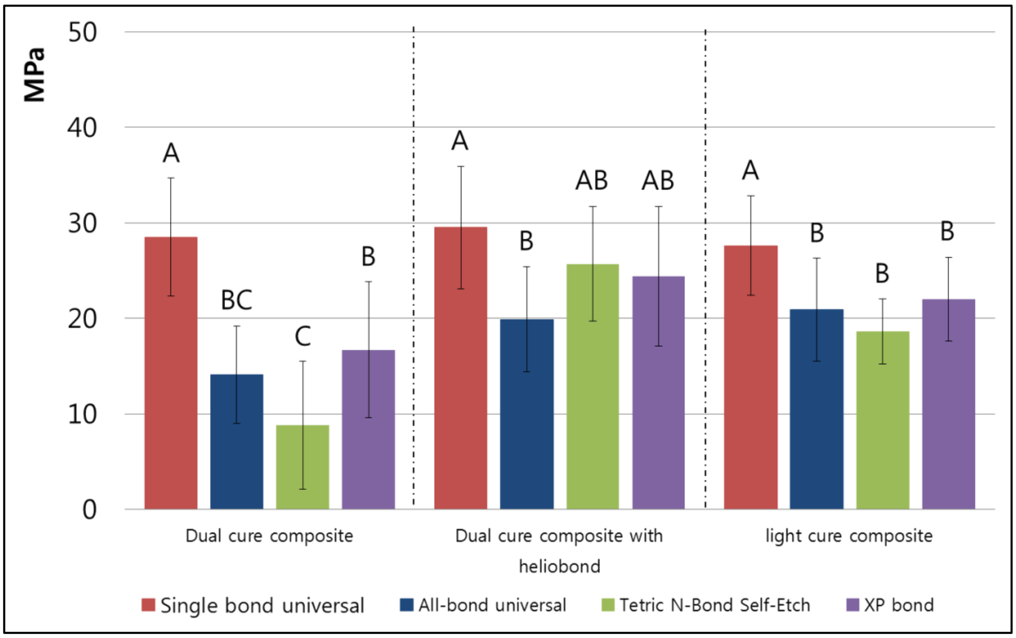


Fig. 3. Mean microshear bond strength values (MPa) of adhesives. Identical letters indicate that values are not significant different with Tukey's test ($p < 0.05$).

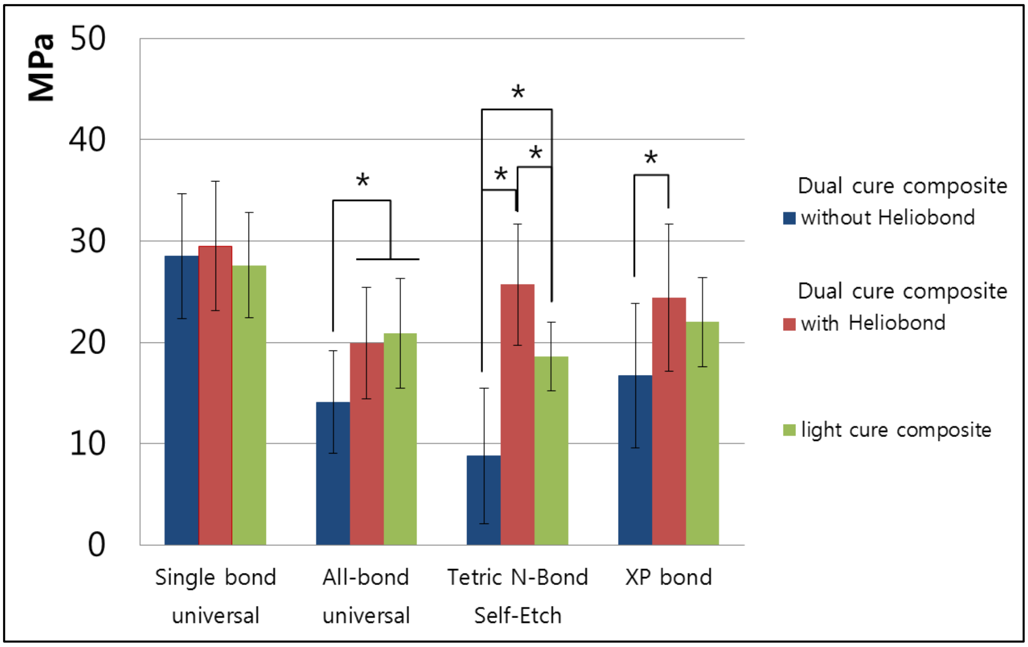


Fig. 4. Mean microshear bond strength values (MPa) with hydrophobic resin coat and different composites. Significant differences are indicated by asterisk (* : $p < 0.05$).

2. Surface evaluation

The distribution of failure modes is shown in Fig. 5 - 7. Single Bond Universal showed a mixed failure – dentin and adhesive layer existed together along with some remaining composite material. In case of other adhesives, high percentages of adhesive failure appeared (Fig. 5). When a hydrophobic resin coat was applied, the groups showed different patterns of failure. Single Bond Universal and All-Bond Universal did not show much difference, Whereas Tetric N-bond Self-etch and XP bond showed an increased percentage of mixed failure (Fig. 6). SEM images of representative fractured surface in each group are shown in Fig. 8 - 13

Fig. 5. Distribution of failure modes (%) with dual-cured composite

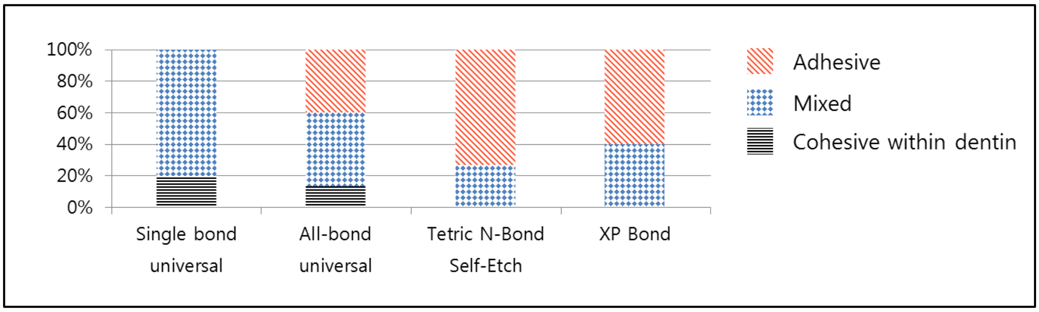


Fig. 6. Distribution of failure modes (%) with dual-cured composite under the application of a hydrophobic resin coat

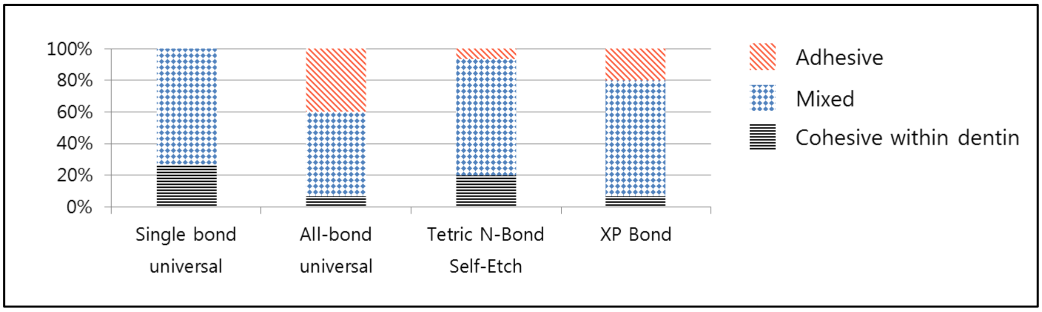
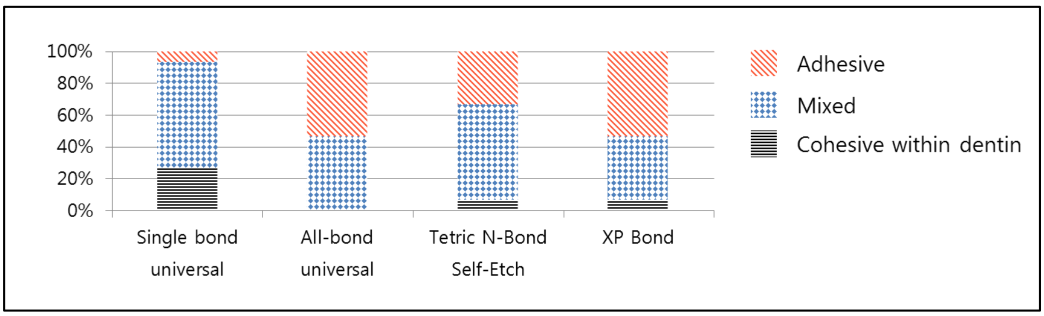


Fig. 7. Distribution of failure modes (%) with light-cured composite



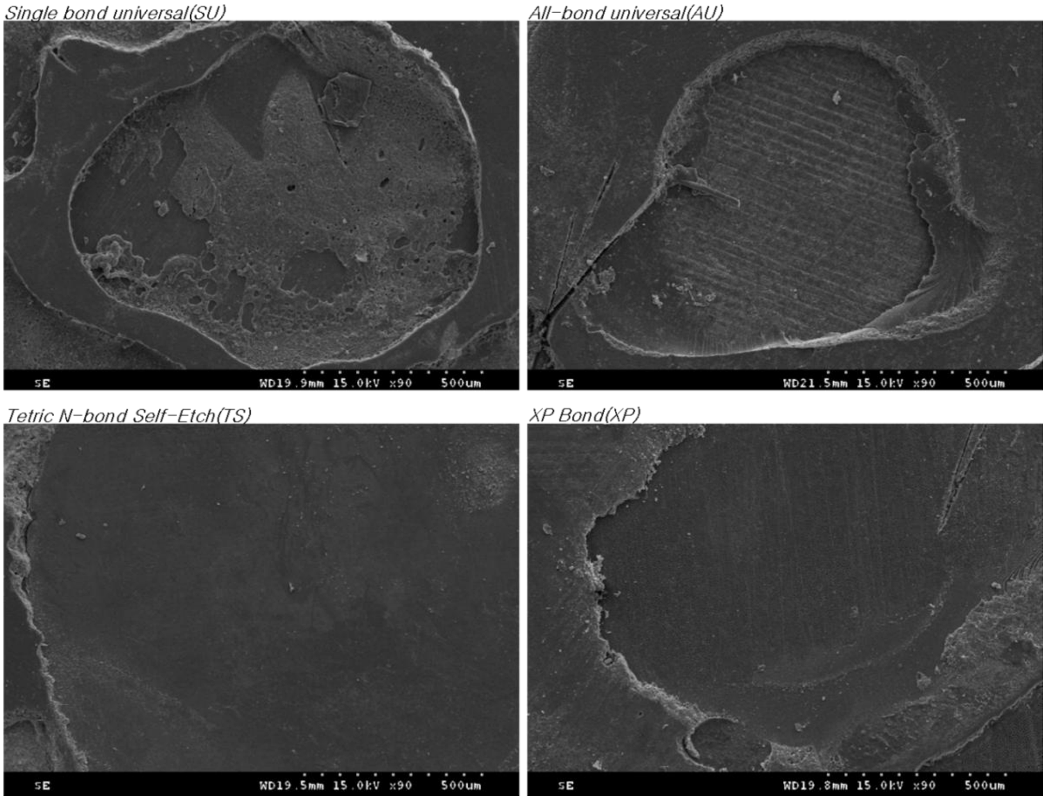


Fig. 8. SEM photomicrographs (X90) of fractured surfaces. SU: mixed failure including the adhesive layer, dentin and partially some composite fragments. AU: adhesive failure between the adhesive and dentin. TS: adhesive failure between the adhesive and composite. XP: adhesive failure between the adhesive and dentin

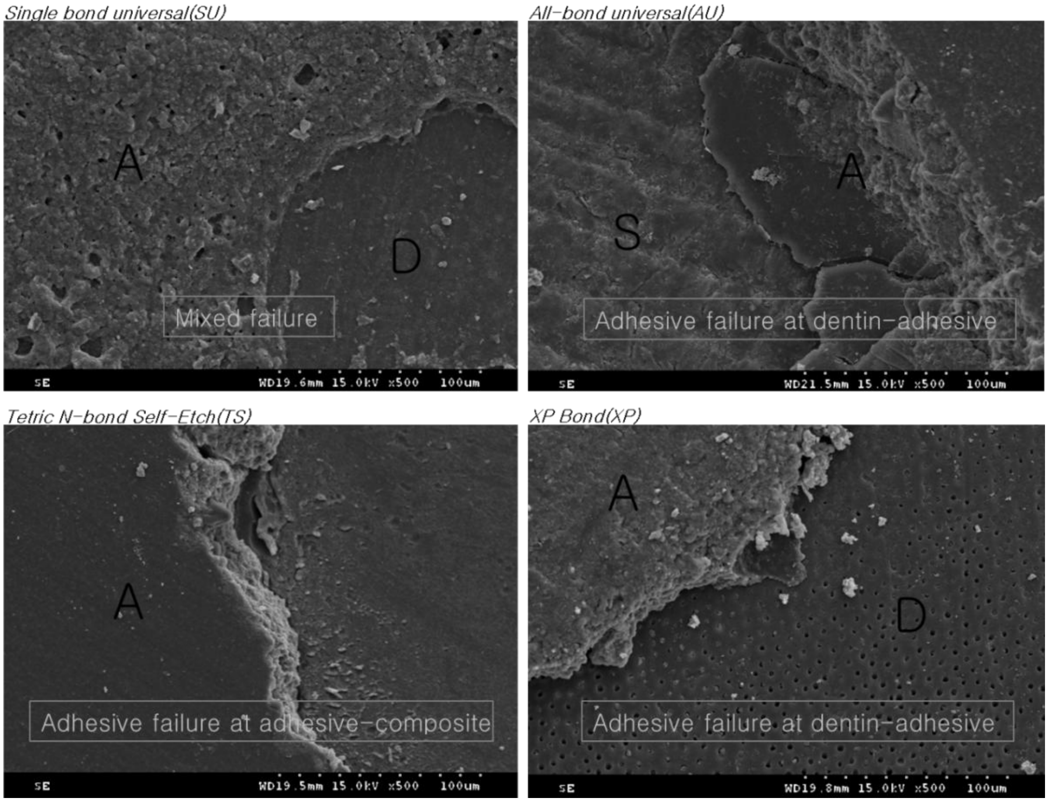


Fig. 9. SEM photomicrographs (X500) showing magnifications of border lines at the fractured surface. SU: dentin (D) and adhesive layer (A) can be observed. AU: a smear layer is present, and the adhesive layer (A) is divided into thinner layers. TS: although some defects exist in the surface, a generally smooth adhesive layer (A) can be found. XP: open dentinal tubules can be seen in the dentin (D) layer, with dentinal tubules closed with resin tags in the periphery.

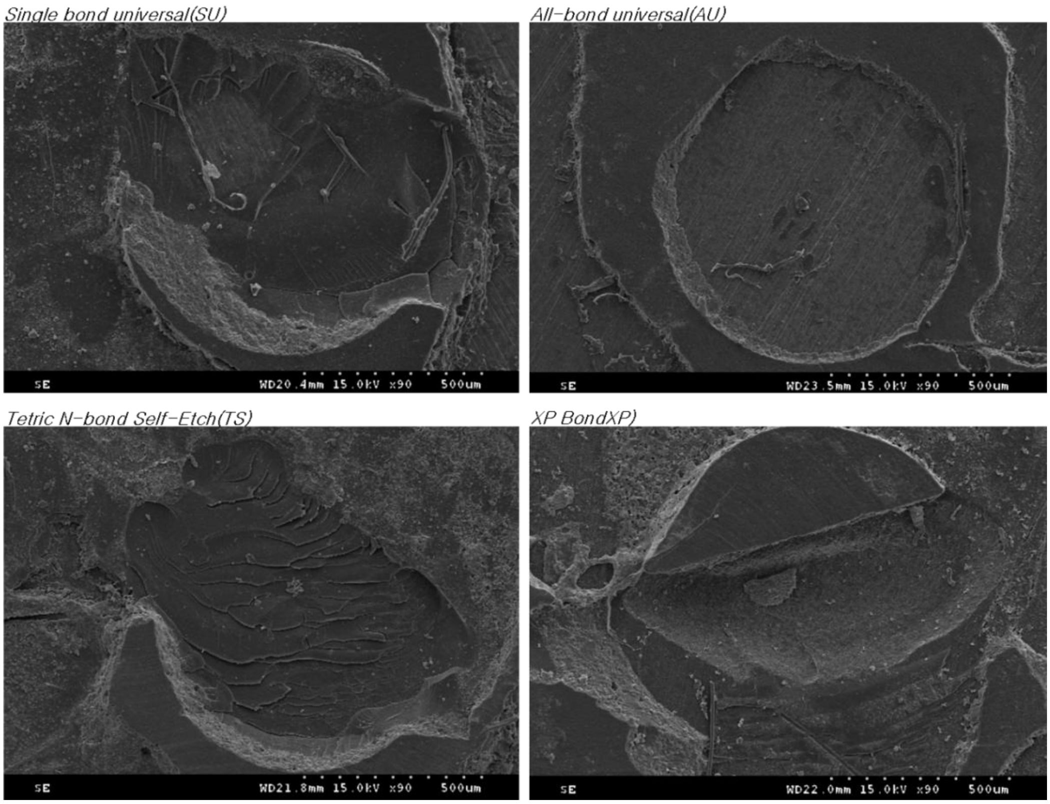


Fig. 10. SEM photomicrographs (X90) of fractured surface of groups with the application of a hydrophobic resin layer. SU: a mixed failure, showing different layers in the surface. AU: the adhesive clearly separated from the dentin surface, as in the test without the hydrophobic resin layer. TS: a mixed failure, with a partially torn mixed adhesive layer. XP: mixed failure with fracture including some dentin and adhesive layer with some clearly remaining composite.

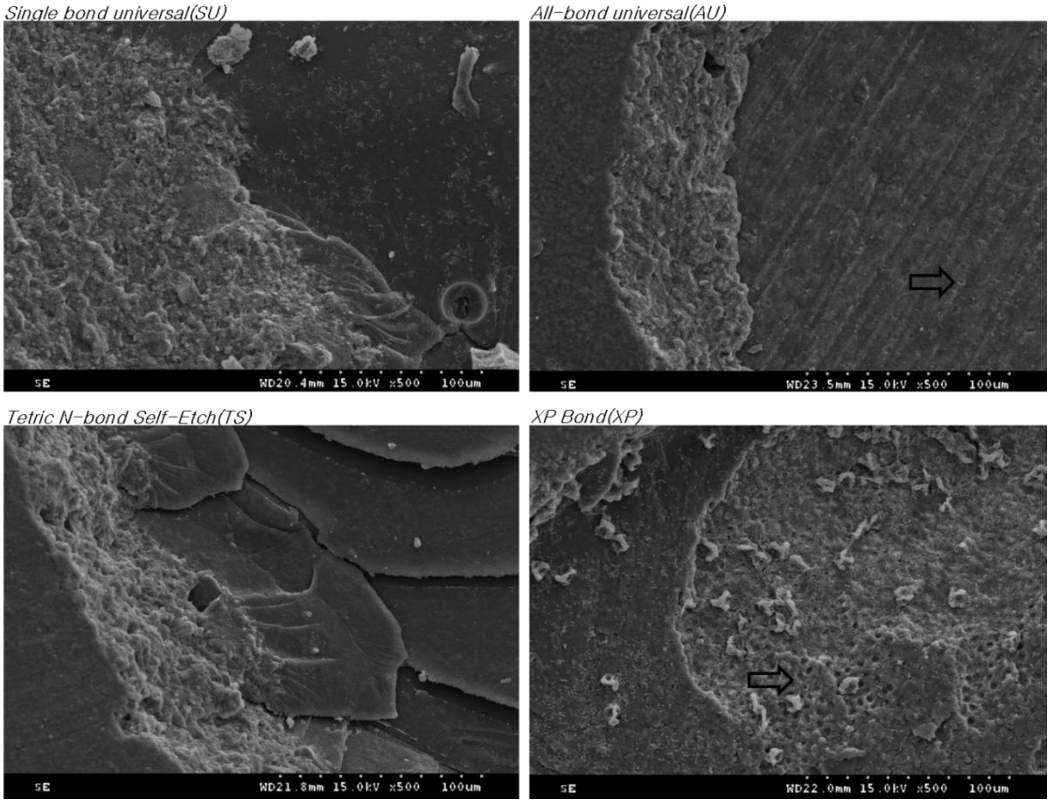


Fig. 11. SEM photomicrographs (X500) showing magnifications of border lines at the fractured surface in hydrophobic resin layer groups. AU: adhesive failure between the adhesive and dentin. A uniform smear layer exists in the dentin layer with some signs of dentinal tubules (arrow). XP: Various layers can be observed, with an evident fracture defect in the dentin layer including clear signs of dentinal tubules (arrow).

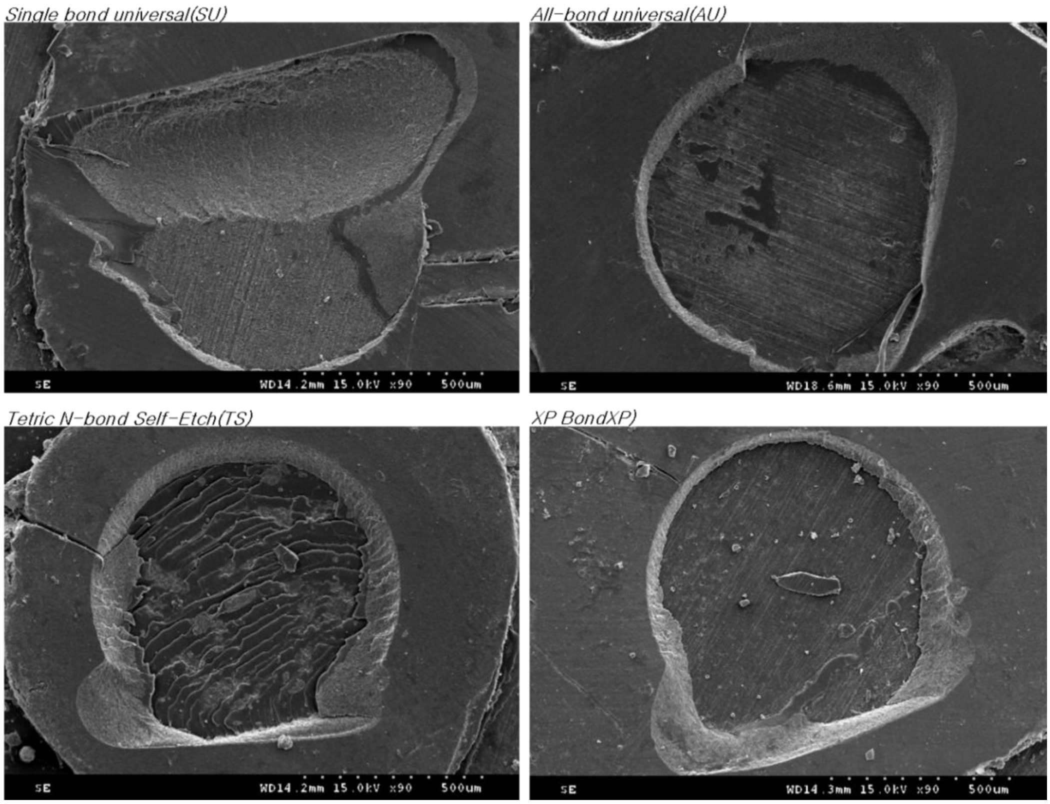


Fig. 12. SEM photomicrographs (X90) of fractured surface of control groups (light-cured composite). SU: a mixed failure, showing a large area of cohesive failure in the dentin with some residual adhesive. AU: adhesive failure between the adhesive and dentin. TS: a mixed failure, with dentin exposed in several areas with the adhesive torn into layers. XP: adhesive failure between the adhesive and dentin.

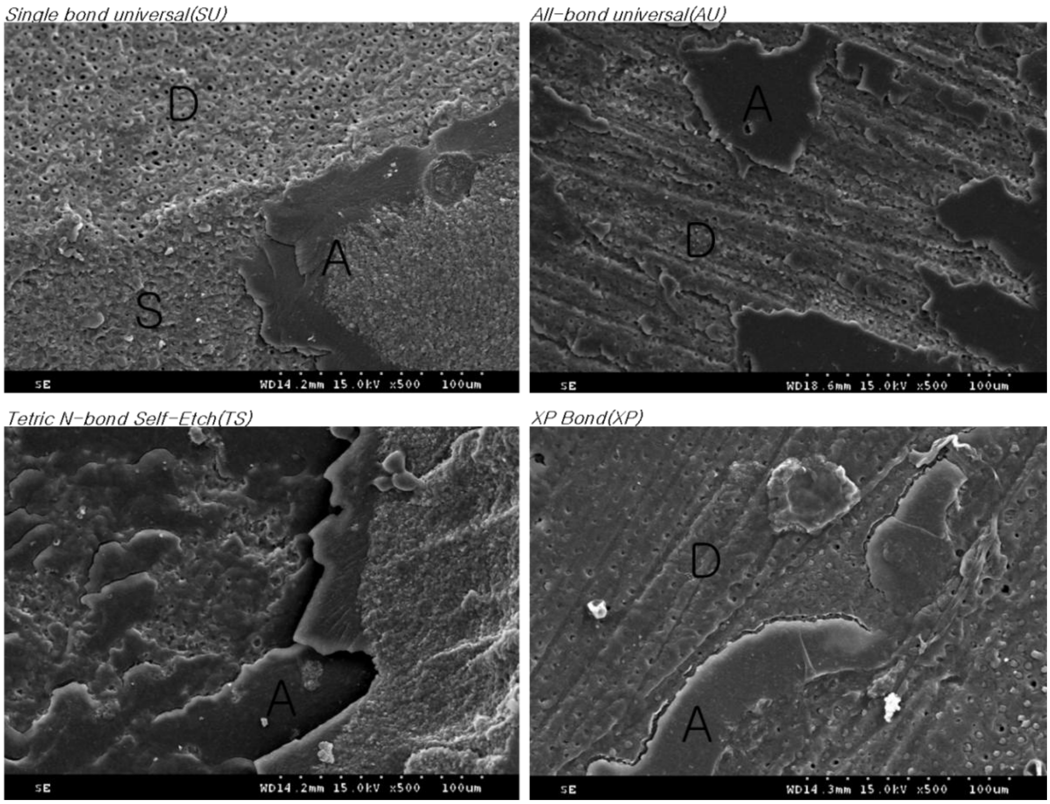


Fig. 13. SEM photomicrographs (X500) showing magnifications of border lines at the fractured surface of control groups (light-cured composite). SU: the adhesive layer (A) and smear layer (S) can be observed, with dentinal tubules clearly exposed due to cohesive fracture in the dentin (D). AU: residual adhesive (A) fragments and dentinal tubules around the smear layer can be found in the dentin (D). TS: adhesive found in different layers with the smear layer and some dentinal tubules showing beneath. XP: dentinal tubules are apparent in the dentin (D) with some adhesive (A) fragments.

IV. Discussion

The incompatibility of dual-cured composite with one-step self-etch adhesive was mentioned previously (Swift et al., 1998). Especially self-cured composite resin with one-step self-etch adhesives showed strong unsuitability (Shade et al., 2014). A study by Giannini et al. on the compatibility of dual-cured composite with one-step self-etch adhesive – through light activation and chemical activation – showed weak bonding strength even with direct light activation (Giannini et al., 2004). Furthermore, a previous study demonstrated that light-cured resins are not totally immune to adverse acid-base reactions when a tertiary amine is unutilized as an accelerator for the camphorquinone photosensitizer (Suh et al., 2003). This shows that even with light curing, dual-cured composite is still incompatible with one-step self-etch adhesives. Moreover, even during clinical procedures, incremental filling and light curing are more advantage and widely used. Therefore light curing was preferred to self-curing to test the incompatibility with dual-cured composite. In present study, LuxaCore® Dual was used as dual-cured composite to experiment its compatibility with simplified step adhesives. After filling, an immediate 20-second light curing procedure was undertaken.

The reason of incompatibility of one-step self-etch adhesive and dual-cured/self-cured composite is the acidic (Schittly et al., 2010) and hydrophilic (Chen and Suh, 2013) nature of the adhesive. However, according to the manufacturers, recently introduced new universal adhesives (e.g. All-Bond Universal, Bisco) are expected to improve the incompatibility with dual-cured composite via the mild acidity (pH>3) and increase in hydrophobicity. In present study, Tetric N-Bond Self-Etch, a one-step self-etch system agent, demonstrated the weakest bonding strength and showed incompatibility with dual-cured composite similar to previous studies (Swift et al., 1998; Tay et al., 2003a). However, regarding recently introduced universal systems, Single Bond Universal and All-Bond Universal, All-Bond Universal showed no significant difference with Tetric N-Bond Self-Etch, but Single Bond Universal showed the highest bonding strength under simplified application with the conventional one- step self-etch system. In comparison to All-Bond Universal, Single Bond Universal showed noteworthy difference concerning bonding strength. This suggests that there might be a notable difference regarding the compatibility with dual-cured composite. Both agents include HEMA and MDP, a phosphate monomer, and ethanol and water as solvents. MDP, an acidic resin monomer that can affect the degree of incompatibility with dual-cured composite, is included in both of the bonding agents. However, All-Bond Universal and Single bond show varying results concerning the incompatibility with dual-cured composite. Single Bond Universal includes additives such as Vitrebond™ Copolymer, filler and silane which not only influence the bonding strength, but also affect the ratio of MDP in the agent. Alike MDP, Vitrebond™ Copolymer, which is a polyalkenoic acid copolymer, provides a chemical bond with dentin substrate (Perdigao et al., 2014). Thus, it can be expected that Vitrebond™ Copolymer enables Single Bond Universal to have an increased compatibility with dual-cured composite and a more efficient adhesion with dentin substrate while decreasing the ratio of MDP. Whether the percentage of MDP influences the compatibility with dual-cured composite is to be further studied.

Moreover, Single Bond Universal showed the highest bonding strength regardless of compatibility. Another possible explanation can be that it includes a specific ratio of filler unlike other adhesives. The addition of filler in to the adhesive can augment the bonding strength by decreasing polymerization shrinkage and strengthening the hybrid layer (Faltermeier et al., 2007; Gallo et al., 2001). The results with Single Bond Universal are congruent with other recent studies showing that higher bond strength was obtained with Single Bond Universal among other one-step self-etching adhesives (Munoz et al., 2013; Taschner et al., 2013).

To study the incompatibility of simplified etch and rinse system and dual-cured composite, XP bond was also included in the test group. Tay et al. reported an incompatibility between dual-cured composite and single-bottle, total-etch adhesive system, and advised when using on hydrated dentin, the permeability of the adhesive is the key factor influencing the compatibility (Tay et al., 2003a; Tay et al., 2003b). However, this in-vitro study used dentin slices, and consequently the results will be different to those that dealt with vital teeth, naturally affected by dentinal fluid. Permeability is a less critical factor in this experiment.

Another influence on the incompatibility would be pH. In a research on the incompatibility between single-bottle, total-etch adhesive system and dual-cured composite, Sanares et al. mentioned a notable relationship between the pH of the adhesive and bonding strength (Sanares et al., 2001). A lower pH resulted in a weaker bonding strength. Thus, it can be predicted that the low pH of XP bond (pH=2.5) affected the incompatibility as with other one-step self-etch systems (according to the manufacturer's instructions, Single Bond Universal pH=2.7; All-Bond Universal pH=3; Tetric N-Bond Self-Etch pH=1.5).

Unlike manufacturers' information, Munoz et al. reported different pH of the adhesives: Single Bond Universal pH = 3, All-Bond Universal pH = 2.4 (Munoz et al., 2013). Thus, it is necessary to test for the pH of the applied adhesives and interpret the results of this experiment with the tested information. The present study utilized colorpHast® (EMD, Germany) to test the pH of the adhesive. In our results, pH values were measured to be different to the manufacturer's information – All-Bond Universal was measured pH 4 (slightly higher than pH 3), and XP bond was pH 4.5 (a much higher value than 2.5). In case of All-Bond Universal, its high pH will affect the bond strength with dentin when applied using the self-etch system. According to Suyama et al.'s study, the higher the pH of the self-etch adhesives, the lower the bond strengths with dentin (Suyama et al., 2013). This is because the pH of adhesives lays an effect on the etching ability of dentin which includes a smear layer. Regardless of the incompatibility with dual-cured composite, All-Bond Universal, with a higher pH, showed lower bond strength compared to Single Bond Universal in this study. This is since unlike Single Bond Universal, which mostly showed a mixed failure, All-Bond Universal showed a clear adhesive failure between the dentin and adhesive layer. This was supported by the SEM images showing the remnant smear layer in the dentin surface. Also, the application of a hydrophobic resin coat did not modify the failure mode with dual-cured composite, and this result is thought to be due to the low bond strength between the dentin and adhesive rather than a mere incompatibility between the agents. On the other hand, the pH of XP bond was measured to be higher than its label value, and this would have made a positive effect on its compatibility with dual-cured composite. This is supported by the results showing similar bond strengths with dual-cured composite and light-cured composite.

As mentioned earlier, the incompatibility of dual-cured composites and self-etch adhesives usually exists when a low-pH adhesive is used before applying the composite resin (Schittly et al., 2010; Tay et al., 2003a). The unsuitable reaction occurs between the acidic resin monomer in the unpolymerized acidic adhesive layer and tertiary amine, a polymerization catalyst of composite resin. The presence of these ‘left-over’ acidic resin monomers on the surface of the adhesive is created as an oxygen inhibition layer forms with contact with oxygen during polymerization. In this experiment all groups except for Single Bond Universal showed a marked increase in bond strength when an additional hydrophobic resin coat was applied. It is expected that this extra hydrophobic resin coat acts as an intermediate layer that helps avoid the unpolymerized acidic resin monomer from coming in contact with the tertiary amine of composite resin. In fact a lot of researches have reported that 2-step self-etch system with hydrophobic adhesives show higher bonding strength and better compatibility with composite resin than 1-step self-etch system (Giannini et al., 2004; Munoz et al., 2013; Reis et al., 2009). Our results did not show the difference of microshear bond strength among the adhesives after applying hydrophobic resin layers. That is, after the application of the hydrophobic resin coats, the adhesives showed improved bond strength to a similar extent compared to the control group. This result is in accordance with that of another study that evaluated bond strength with a hydrophobic resin coat, which revealed that the use of a hydrophobic resin coat significantly improves the degree of conversion in dentin (J Perdigao, 2014). Additionally, in groups which showed weak bond strength and higher rates of adhesive failure without the application of a hydrophobic resin coat in the failure mode analysis of this study, the bond strength and ratio of mixed failure increased with the application of a hydrophobic resin coat. SEM images also showed mixed failure with the presence of a hydrophobic resin coat. Overall, the present study showed a higher tendency for mixed failures between the adhesive and dentin with higher microshear bond strength and adhesive failures between with lower microshear bond strength. But the interpretation of failure mode remains challenging.

Unlike other test groups, the Single Bond Universal group showed excellent bonding strength regardless of an additional hydrophobic resin layer. A recent 18-month clinical trial has reported scarcely any difference with the application of an extra hydrophobic resin layer in Adper Easy Bond (3M ESPE), a 1 step self-etch system agent (Sartori et al., 2013). Similar results were also obtained in a 18-month clinical evaluation using Single Bond Universal (Perdigao et al., 2014). These researches mentioned that the two adhesives include a similar ratio of polyalkenoic acid copolymers, and that the influence of the hydrophobic resin layer depends on a specific component of the one-step self-etch agent (Perdigao et al., 2014; Sartori et al., 2013).

In the present study, previous to testing the incompatibility between dual-cured composite and simplified step adhesive, light-cured composite was utilized and the microshear bond strength tests were carried out as a control group to investigate the bond strength of the adhesives and their effects. As the control, light-cured composite which showed the most similar flexural strength (≒120MPa) as the tested dual-cured composite was used. When applying the light-cured composite, the Tetric N-Bond Self-Etch group showed higher bond strength compared to dual-cured composite, which implies the incompatibility with dual-cured composite. All-Bond Universal also showed significantly higher bond strength with the application of light-cured composite than dual-cured composite. Moreover, the bond strength with light-cured composite was similar to that of dual-cured composite with the application of a hydrophobic resin coat – this suggests that the incompatibility between All-Bond Universal and dual-cured composite still remains. However, various factors can affect the bond strength and the evaluation of incompatibility limited to these factors is not absolutely sufficient. Thus, cautious interpretation is necessary.

According to Marshall et al., various factors affect the bonding strength, and those related to the tooth matrix itself include the depth, direction and location of dentin, presence of dental caries as well as age (Marshall et al., 1997). Thus, by utilizing dentin of equal depths and directions an effort was made to standardize the slices. The composite resin specimens produced in this test had a small surface area with a diameter of 0.7mm. Sano et al. showed that as the bonding area decreases, the bonding strength increases exponentially (Sano et al., 1994) – that is, the increase in bonding area will weaken the bonding strength. This is because the bonding surface does not show a uniform pattern and may contain air resulting in separation of the bonding surface or show irregular film thickness. Due to the irregular roughness of the surface, stress is distributed unevenly; and a larger bonding surface area would imply the presence of more defects in the surface.

Thus, 0.7mm-diameter composite resin specimens with small surface areas were considered to be apt for this experiment.

Microshear bond tests were carried out to compare bonding strengths. Based on finite element stress analysis, shear force usually shows an intrinsic limitation – as stress is not distributed in the bonding surface but is concentrated at the base, cohesive failure occurs at the base of the material (Armstrong et al., 2010). Taking this into account, a microtensile test could be a more suitable method, but this procedure also carries the possibility of cohesive failure (Scherrer et al., 2010), and is very technique sensitive to produce the specimen (Shimada et al., 2002). The shear bond test, on the other hand, would be an appropriate method to evaluate the compatibility between the materials. Because there is no need for trimming while producing the specimens, it is less likely to drop or destroy the specimen during the fabrication process, and weak bonding strengths can be also be measured in specimens with incompatible materials. In fact, several researches on the compatibility of materials have planned and utilized shear bond tests (Chen and Suh, 2013; Giannini et al., 2004; Ishii et al., 2008; Roh and Chung, 2005; Schittly et al., 2010).

The limitation of this study was that bond strengths by microshear bond tests were used to evaluate the compatibility between materials. Experiments with other designs of conditions could demonstrate different results. Moreover, a further evaluation of the exact composition ratio of each adhesive and the influence of each component would be necessary. Since the bond strength of dual-cured composite varied depending on the type of adhesive, the tested null hypothesis was not supported in this study. Likewise, except for the Single Bond Universal group, the bond strength increased with the application of a hydrophobic resin layer, and this result also denies the null hypothesis.

V. Conclusion

The aim of this article was 1) to evaluate the influence of two different universal adhesive on micro shear bond strength of dual-cured composite to dentin, 2) to evaluate the effect of application of a hydrophobic resin coat on the bond strength within the hybrid layer of a one-step self-etch adhesive system.

Within the limitation of this study, the following conclusions were drawn:

(1) With respect to the influence of universal adhesives, Single Bond Universal showed the highest microshear bond strength but not incompatibility with dual-cured composite ($p < 0.0001$). However All-Bond Universal showed relatively low microshear bond strength and statistically no difference with Tetric N-Bond Self-Etch as existing one-step self-etch system.

(2) Hydrophobic resin coat may be considerable alternation for solving the problem of incompatibility between single step adhesive system and dual-cured composite, and can improve microshear bond strength of these materials.

References

Armstrong S, Geraldeli S, Maia R, Raposo LH, Soares CJ, Yamagawa J: Adhesion to tooth structure: a critical review of "micro" bond strength test methods. Dent Mater 26(2): e50-62, 2010.

Chen L, Suh BI: Effect of hydrophilicity on the compatibility between a dual-curing resin cement and one-bottle simplified adhesives. J Adhes Dent 15(4): 325-331, 2013.

Faltermeier A, Rosentritt M, Faltermeier R, Reicheneder C, Mussig D: Influence of filler level on the bond strength of orthodontic adhesives. Angle Orthod 77(3): 494-498, 2007.

Gallo JR, Comeaux R, Haines B, Xu X, Burgess JO: Shear bond strength of four filled dentin bonding systems. Oper Dent 26(1): 44-47, 2001.

Giannini M, De Goes MF, Nikaido T, Shimada Y, Tagami J: Influence of activation mode of dual-cured resin composite cores and low-viscosity composite liners on bond strength to dentin treated with self-etching adhesives. J Adhes Dent 6(4): 301-306, 2004.

Haller B: Which self-etch bonding systems are suitable for which clinical indications? Quintessence Int 44(9): 645-661, 2013.

Imai Y, Kadoma Y, Kojima K, Akimoto T, Ikakura K, Ohta T: Importance of polymerization initiator systems and interfacial initiation of polymerization in adhesive bonding of resin to dentin. J Dent Res 70(7): 1088-1091, 1991.

Inoue S, Koshiro K, Yoshida Y, De Munck J, Nagakane K, Suzuki K, et al.: Hydrolytic stability of self-etch adhesives bonded to dentin. J Dent Res 84(12): 1160-1164, 2005.

Ishii T, Ohara N, Oshima A, Koizumi H, Nakazawa M, Masuno T, et al.: Bond strength to bovine dentin of a composite core build-up material combined with four different bonding agents. J Oral Sci 50(3): 329-333, 2008.

King NM, Tay FR, Pashley DH, Hashimoto M, Ito S, Brackett WW, et al.: Conversion of one-step to two-step self-etch adhesives for improved efficacy and extended application. Am J Dent 18(2): 126-134, 2005.

Marshall GW, Jr., Marshall SJ, Kinney JH, Balooch M: The dentin substrate: structure and properties related to bonding. J Dent 25(6): 441-458, 1997.

Moll K, Schuster B, Haller B: Dentin bonding of light- and self-curing resin composites using simplified total- and self-etch adhesives. Quintessence Int 38(1): e27-35, 2007.

Munoz MA, Luque I, Hass V, Reis A, Loguercio AD, Bombarda NH: Immediate bonding properties of universal adhesives to dentine. J Dent 41(5): 404-411, 2013.

Nyunt MM, Imai Y: Adhesion to dentin with resin using sulfinic acid initiator system. Dent Mater J 15(2): 175-182, 1996.

Pashley DH, Sano H, Ciucchi B, Yoshiyama M, Carvalho RM: Adhesion testing of dentin bonding agents: a review. Dent Mater 11(2): 117-125, 1995.

Perdigao J: New developments in dental adhesion. Dent Clin North Am 51(2): 333-357, viii, 2007.

Perdigao J, Kose C, Mena-Serrano AP, De Paula EA, Tay LY, Reis A, et al.: A new universal simplified adhesive: 18-month clinical evaluation. Oper Dent 39(2): 113-127, 2014.

Perdigao J, Munoz M, Sezinando A, Luque-Martinez I, Staichak R, Reis A, et al.: Immediate Adhesive Properties to Dentin and Enamel of a Universal Adhesive Associated With a Hydrophobic Resin Coat. Oper Dent, 2013.

Reis A, Leite TM, Matte K, Michels R, Amaral RC, Geraldeli S, et al.: Improving clinical retention of one-step self-etching adhesive systems with an additional hydrophobic adhesive layer. J Am Dent Assoc 140(7): 877-885, 2009.

Roh OD, Chung JH: Micro-shear bond strength of five resin-based composites to dentin with five different dentin adhesives. Am J Dent 18(6): 333-337, 2005.

Sanares AM, Itthagarun A, King NM, Tay FR, Pashley DH: Adverse surface interactions between one-bottle light-cured adhesives and chemical-cured composites. Dent Mater 17(6): 542-556, 2001.

Sano H, Shono T, Sonoda H, Takatsu T, Ciucchi B, Carvalho R, et al.: Relationship between surface area for adhesion and tensile bond strength--evaluation of a micro-tensile bond test. Dent Mater 10(4): 236-240, 1994.

Sartori N, Peruchi LD, Guimaraes JC, Silva SB, Monteiro S, Jr., Baratieri LN, et al.: Clinical effectiveness of a hydrophobic coating used in conjunction with a one-step self-etch adhesive: an 18-month evaluation. Oper Dent 38(3): 249-257, 2013.

Scherrer SS, Cesar PF, Swain MV: Direct comparison of the bond strength results of the different test methods: a critical literature review. Dent Mater 26(2): e78-93, 2010.

Schittly E, Bouter D, Le Goff S, Degrange M, Attal JP: Compatibility of five self-etching adhesive systems with two resin luting cements. J Adhes Dent 12(2): 137-142, 2010.

Shade A, Wajdowicz M, Bailey C, Vandewalle K: The Effect of Simplified Adhesives on the Bond Strength to Dentin of Dual-cure Resin Cements. Oper Dent, 2014.

Shimada Y, Yamaguchi S, Tagami J: Micro-shear bond strength of dual-cured resin cement to glass ceramics. Dent Mater 18(5): 380-388, 2002.

Suh BI, Feng L, Pashley DH, Tay FR: Factors contributing to the incompatibility between simplified-step adhesives and chemically-cured or dual-cured composites. Part III. Effect of acidic resin monomers. J Adhes Dent 5(4): 267-282, 2003.

Suyama Y, Luhrs AK, De Munck J, Mine A, Poitevin A, Yamada T, et al.: Potential smear layer interference with bonding of self-etching adhesives to dentin. J Adhes Dent 15(4): 317-324, 2013.

Swift EJ, Jr., May KN, Jr., Wilder AD, Jr.: Effect of polymerization mode on bond strengths of resin adhesive/cement systems. J Prosthodont 7(4): 256-260, 1998.

Taschner M, Kummerling M, Lohbauer U, Breschi L, Petschelt A, Frankenberger R: Effect of Double-layer Application on Dentin Bond Durability of One-step Self-etch Adhesives. Oper Dent, 2013.

Tay FR, Pashley DH, Yiu CK, Sanares AM, Wei SH: Factors contributing to the incompatibility between simplified-step adhesives and chemically-cured or dual-cured composites. Part I. Single-step self-etching adhesive. J Adhes Dent 5(1): 27-40, 2003a.

Tay FR, Suh BI, Pashley DH, Prati C, Chuang SF, Li F: Factors contributing to the incompatibility between simplified-step adhesives and self-cured or dual-cured composites. Part II. Single-bottle, total-etch adhesive. J Adhes Dent 5(2): 91-105, 2003b.

국문 요약

새로운 universal 상아질 접착제의 이원 중합형 복합 레진에 대한 적합성 및 hydrophobic resin coat의 영향

(지도 교수 노 병 덕)

연세대학교 대학원 치의학과

정 건 석

1. 서론

최근 소개되고 있는 universal 상아질 접착제는 다양한 재료와의 호환성을 지니고 있다. 특히 기존에 알려진 재료간의 부적합성(all-in-one타입의 상아질접착제와 화학/이원 중합형 복합 레진)이 개선되었을 것으로 기대된다. 이에 본 연구에서는 이원 중합형 복합 레진의 상아질에 대한 미세 전단 접착 강도에 서로 다른 두 가지 universal 상아질 접착제의 영향에 대해 평가하고, hydrophobic resin coat가 접착 강도에 미치는 효과 역시 평가하고자 하였다.

2. 본론

Universal 상아질 접착제인 Single Bond Universal과 All-Bond universal, one-step self-etch system의 Tetric N-Bond Self-Etch 그리고 etch & rinse system의 XP bond, 총 4가지 상아질 접착제와 hydrophobic 상아질접착제인 Heliobond를 사용하였다. 이원 중합형 복합 레진과 함께 대조군으로 광중합형 복합 레진도 함께 사용하였다. 각각의 상아질 접착제를 사용하여 복합 레진을

상아질시편에 접착 후 만능시험기를 이용하여 미세 전단 접착 강도 (MPa)를 측정하였고, 광학현미경으로 파절면을 관찰하여 파절 양상을 기록하였다. 추가적으로 주사 전자 현미경으로 파절된 계면을 자세히 관찰하였다.

실험 결과, 이원 중합형 복합 레진 이용 시 Single Bond Universal은 통계적으로 유의하게 가장 높은 접착 강도를 보였으나, All-Bond Universal은 낮은 접착강도를 보이며 Tetric N-Bond Self-Etch와 XP bond에 유의한 차이를 나타내지 않았다. 파절 양상도 mixed failure가 주로 관찰된 Single Bond Universal과는 달리 All-Bond Universal에서는 adhesive failure가 주를 이루었다. Hydrophobic resin coat를 적용 시, 대조군과 비슷한 정도의 접착강도를 보였다. Single Bond Universal을 제외한 모든 군에서 통계적으로 유의한 차이가 있는 접착 강도의 증가를 보였고 파절 양상도 mixed failure의 비율이 증가하였다.

3. 결론

(1) Single Bond Universal은 가장 높은 미세 전단 접착 강도를 나타냈으며 이원 중합형 복합 레진과 우수한 호환성을 보였다. 그러나 All-Bond Universal은 낮은 미세 전단 접착 강도를 보였고 Tetric N-Bond Self-Etch와 통계적 유의한 차이를 보이지 않았다.

(2) Hydrophobic resin coat는 상아질 접착제의 미세 전단 접착 강도를 향상 시킬 수 있었으며, 이원 중합형 복합 레진과의 부적합성을 개선하기 위한 대안이 될 수 있을 것이다.

핵심 되는 말: 미세 전단 접착 강도, universal 상아질 접착제, 자가부식 상아질 접착제, 이원 중합형 복합 레진, 적합성, 부적합성, hydrophobic resin coat