상아질 접착제의 접착층 두께가 복합 레진과 상아질 간의 미세인장 접착강도에 미치는 영향

권태엽[1], 김영진[2*]

경북대학교 치의학전문대학원 치과생체재료학교실 및 경북대학교 생체재료연구소[1],
경북대학교 치의학전문대학원 소아치과학교실[2]

⟨Abstract⟩

Influence of Adhesive Layer Thickness of Dentin Adhesives on Microtensile Bond Strength of a Resin Composite to Dentin

Tae-Yub Kwon[1], Young-Jin Kim[2]*

Department of Dental Biomaterials, School of Dentistry; and Institute for Biomaterials Research & Development; Kyungpook National University[1],
Department of Pediatric Dentistry, School of Dentistry, Kyungpook National University[2]

상아질 접착제의 올바른 선택과 사용은 성인 뿐 아니라 소아 환자에서 신속하고 정확한 레진 수복에 있어서 매우 중요하다. 본 연구에서는 상아질 접착제의 본딩 레진의 두께가 복합 레진의 상아질에 대한 미세인장 접착강도에 미치는 영향을 평가하였다. 상아질 접착제로는 All-Bond 2와 All-Bond 3를 사용하였다. 본딩 레진의 두께를 50, 100, 200 ㎛으로 조절하기 위해서, 해당 두께를 가진 접착성 테이프를 연마된 상아질 면에 접착시켰고, 그 외의 접착 단계는 제조자의 지시를 준수하였다. 접착된 시편은 24시간 37℃ 증류수에 보관하였고, 다이아몬드 쏘를 이용하여 미세인장 접착용 시편들을 제작한 다음 (접착면적 : 0.8 mm^2 (±0.1)) 만능시험기를 이용하여 0.5 mm/min의 속도로 시편을 파절시켜 미세인장 접착강도를 MPa 단위로 구하였다. 실험 결과 All-Bond 2에서는 중간 두께인 100 ㎛에서 가장 높은 접착강도를 나타내었고, 그 다음 200 ㎛이었으며, 50 ㎛에서 가장 낮은 접착강도를 보였다. 한편 All-Bond 3에서는 접착강도가 본딩 레진의 두께에 영향을 받지 않았다. 이는 주로 All-Bond 2의 본딩 레진의 주성분인 2-hydroxyethyl methacrylate(HEMA)의 친수성/소수성과 관련된 것으로 생각된다. 본딩 레진의 두께가 복합 레진의 상아질 접착에 미치는 영향은 접착제에 따라 다르게 나타날 수 있으므로 주의가 필요하다.

Key words : 상아질 접착제, 두께, 미세인장 접착강도, 복합 레진

Ⅰ. INTRODUCTION

While the importance of hybrid layer is emphasized through many studies, less studies are done about the

* Correspondence: 김영진 (ORCID ID: 0000-0003-4085-6105)
(41940) 대구광역시 중구 달구벌대로 2177 경북대학교 치의학전문대학원 소아치과학교실
Tel: +053-600-7221, Fax: +82-53-426-6608
E-mail: yjikim@knu.ac.kr

adhesive layer; that is, the hydrophobic layer overlying the hybrid layer (Kim *et al.* 2014). As the component overlying the hybrid layer, the adhesive layer may help to protect the bonding interface from polymerization shrinkage stresses and act as a stress absorbing layer (Cadenaro *et al.* 2008; Kwon *et al.* 2012).

A number of studies have evaluated the role of the elasticity of the hybrid layer and/or adhesive resin layer

in relieving the polymerization stress of resin composites. Although there seems to be no relationship between the thickness of the hybrid layer and tensile bond strength, concern remains that thin hybrid layers may not provide as much stress-breaking function as thicker hybrid layers (El Zohairy et al. 2005). However, one possible solution is to use thicker adhesive layers on top of thin hybrid layers.

Since there is limited information in the dental literature correlating bond strengths and thickness of the adhesive resin layer, this study evaluated the relationship between the thickness of the adhesive resin layer and microtensile bond strength. The hypothesis is that increased thickness of the adhesive layer would result in higher bond strengths by improving stress distributions in the bonding interface.

II. MATERIALS AND METHODS

Two commercially available dentin adhesives (All-Bond 2 and All-Bond 3, both from Bisco, Inc.) were used. Tetric Ceram (Ivoclar Vivadent) was used as the composite resin. Their manufacturers, lot numbers are summarized in Table 1.

Non-carious human molars were collected after obtaining the patients' informed consents obtained under a protocol approved by the Ethics Committee of the School of Dentistry, Kyungpook National University. The teeth were stored in 0.5% chloramine in water at 4℃ and used within one month following extraction. The occlusal enamel and roots of the teeth were removed using a slow-speed saw with a diamond-impregnated disk (Isomet, Buehler Ltd., Lake Bluff, IL, USA) under water cooling to form 5-6 mm thick, parallel-sided crown segments (Tay et al. 2003). A 600-grit silicon carbide paper was used under running water to create a thin smear layer on the dentin surface (Mazzitelli et al. 2008).

An adhesive tape with a 10 mm diameter hole was placed on the center of the dentin surface to limit the bonding surface area. The three different thickness (50, 100, and 200 μm) of the adhesive layers were controlled by the thickness of the adhesive tape (Table 2). The teeth samples were then randomly divided into six experimental groups (Table 2). The two dentin adhesives used according to the manufacturer's instructions (Table 3) except for the control of the adhesive layer thickness. After applying the bonding resins, the dentin surface was then built up with a resin composite (Tetric Cram, Ivoclar Vivadent) to a height of 5 mm on three increments, light curing each increment for 20 s. All the bonded samples were stored in water at 37℃ for 24 h (Kim et al. 2014).

Each bonded specimen was longitudinally sectioned into

Table 1. Materials used in this study

Materials		LOT #	Manufacturer
All-Bond 2	Etchant	0700006146	Bisco, Inc., Schaumburg, IL, USA
	Primer A	0700003705	
	Primer B	0700005403	
	D/E resin	0700005402	
All-Bond 3	Part A	0700006855	Bisco, Inc., Schaumburg, IL, USA
	Part B	0700006856	
	Bonding resin	0700007053	
Tetric Ceram	A3 shade	GP0300	Ivoclar Vivadent, Schaan, Liechtenstein

Table 2. Six experimental groups

Groups	Dentin adhesive systems	Thickness of the applied adhesive
A1		50 μm
A2	All-Bond 2	100 μm
A3		200 μm
B1		50 μm
B2	All-Bond 3	100 μm
B3		200 μm

Table 3. Instructions according to the manufacturer

Adhesives	Steps	Application methods
All-Bond 2	1	UNI-ETCH (32% phosphoric acid) for 15 s. Rinse and blot-dry.
	2	Apply five consecutive coats of mixed primer. Air-dry. No light curing.
	3	Apply a thin layer of D/E resin. Light curing for 20 s.
All-Bond 3	1	UNI-ETCH (32% phosphoric acid) for 15 sec. Rinse and blot-dry.
	2	Apply 1-2 coats of mixed Parts A and B. Air dry. Light curing for 10 s.
	3	Apply one thin coat of Bonding resin. Light curing for 10 s.

0.9 mm-thick slabs with the slow-speed diamond saw (Isomet). Each slab was fixed on a glass platform with sticky wax and serially sectioned into 0.8 mm^2 (±0.1) sticks, in accordance with the "non-trimming" method of the microtensile test (Mazzitelli *et al.* 2008; Di Hipólito *et al.* 2012). The exact dimensions of each stick were measured using a digital caliper to calculate the precise cross-sectional area. For microtensile bond strength testing, three sticks were sectioned from the center of each bonded specimen (Hiraishi *et al.* 2009). The microtensile bond strength of each bonded specimen was recorded as the average of the three readings (Kim *et al.* 2015). Sticks with premature bond failure were assigned a null bond strength value and were included in the compilation of the mean bond strength as well as the failure mode assessment (Yip *et al.* 2001). The bonded composite-dentin sticks were attached to a testing device with cyanoacrylate glue (Zapit, DVA, Corona, CA, USA). The device was attached to a microtensile bond tester (Bisco Inc.) and loaded in tension at a crosshead speed of 0.5 mm/min until failure (Yip *et al.* 2001).

After fracturing, all specimens were examined under a stereomicroscope (Olympus, Tokyo, Japan) at a magnification of 20× and failure modes were categorized as: (a) adhesive failure along the dentin-resin interface; (b) cohesive failure of the dentin (Cohesive D), and (c) cohesive failure of the resin (Cohesive R) (El Zohairy *et al.* 2005).

The cross-sectional view of the bonding interface was also examined under a field emission-scanning electron microscope (FE-SEM, JSM-6700F, Jeol, Tokyo, Japan). The

specimens were dehydrated through a series of ascending ethanol concentrations (70%, 80%, 95%, three changes in 100%) for 2 hours each, and then left to completely dehydrate in absolute ethanol for an additional 48 hours (Tay *et al.* 2003). Prior to SEM analysis, the specimens were finally air-dried and sputter-coated with platinum (Hiraishi *et al.* 2009).

All the data were statistically analyzed by parametric methods at α = 0.05 because they did not meet the homogeneity of variances assumption (Levene's test). The Kruskal-Wallis test was used to compare the groups, followed by the Mann-Whitney post hoc test, with adjustment of significance levels using the Sidak correction for multiple testing (Yan *et al.* 2010). All the statistical analyses were performed using SPSS 17.0 for Windows (SPSS Inc, Chicago, IL, USA).

III. RESULTS AND DISCUSSION

Figures 1 and 2 show the cross-sectional SEM images of the All-Bond 2 and All-Bond 3-applied groups. According to the photos, the adhesive layers with different thickness, along with the hybrid layer, were well-developed. The formation of the resin tags was also observed.

Table 4 shows the microtensile bond strength values, along with the failure pattern analysis results. The influence of adhesive layer thickness on microtensile bond strength to dentin was found to be material-specific. In the case of All-Bond 2, the relatively thick (100 or 200 μm) adhesive layer produced a superior resin bonding to dentin compared with the thin (50 μm) adhesive layer. In contrast, as regard to All-Bond 3, the thickness of the adhesive layer did not significantly affect the microtensile bond strength.
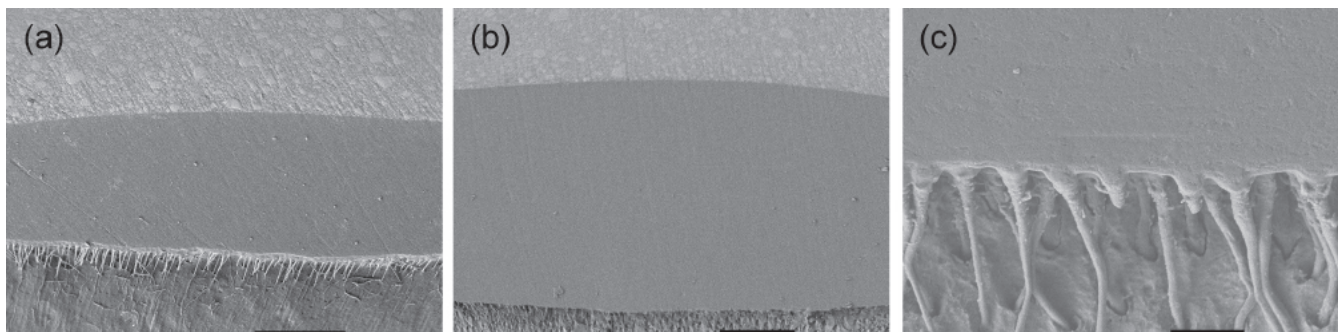
Figure 1. Cross-sectional SEM images of the All-Bond 2-applied groups: (a) 50 μm (250×, bar = 100 μm); (b) 100 μm (200×, bar = 100 μm); and (c) 200 μm (2,000×, bar = 10 μm).
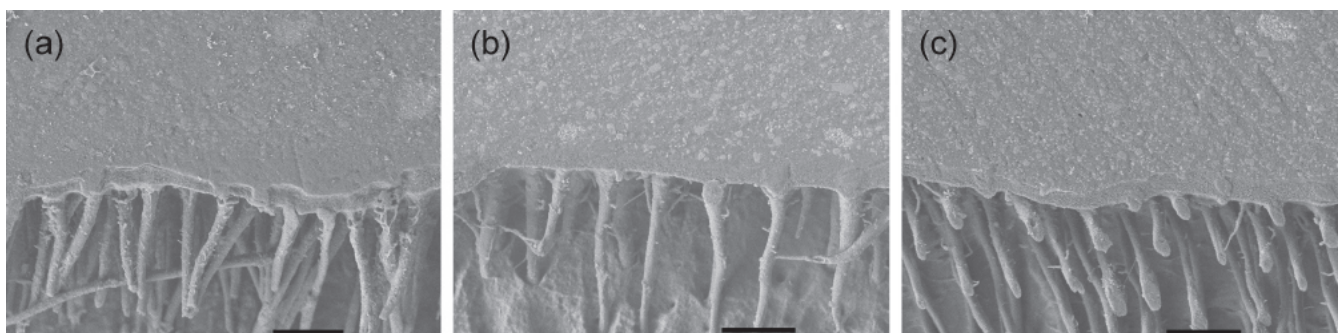
Figure 2. Cross-sectional SEM images of the All-Bond 3-applied groups (2,000×, bar = 10 μm): (a) 50 μm; (b) 100 μm; and (c) 200 μm.

Table 4. Microtensile bond strength and failure pattern of the six experimental groups

Groups	Dentin Adhesive Systems	Microtensile bond strength (MPa, mean ± SD)	Failure pattern
A1		27.4 ± 4.1^b	Adhesive 6; Cohesive R: 2; Cohesive D: 2
A2	All-Bond 2	36.8 ± 8.2^{ab}	Adhesive: 2; Cohesive R: 5; Cohesive D: 3
A3		32.8 ± 6.9^a	Adhesive 4; Cohesive R 3; Cohesive D: 3
B1		21.9 ± 2.8^a	Adhesive 10
B2	All-Bond 3	21.3 ± 5.1^a	Adhesive 9: Cohesive R: 1
B3		20.3 ± 4.3^a	Adhesive 9: Cohesive R: 1

Within an adhesive, the same superscripts are not significantly different ($p \rangle$ 0.05).

These findings may be due to the property or quality of the adhesive layer, rather than that of hybrid layer.

Recent advances in adhesive dentistry have seen a reduction in the application steps of enamel/dentin adhesives to meet the clinical demand for simplified, less technique sensitive bonding procedures (Suh *et al.* 2003; Kwon *et al.* 2012). However, the changes in formulation to develop simplified adhesives entails more acidic, and as a result, more hydrophilic adhesives (Tay & Pashley 2003). As the bonding resin of three-step etch and rinse, and two-step self-etch adhesives, do not contain either acidic monomers or solvents, they form a neutral, relatively hydrophobic adhesive layer prior to the placement of the filling (Kwon *et al.* 2012). On the contrary, two-step etch and rinse, or one-step self-etch adhesives (simplified adhesives), tend to retain a thin, uncured (i.e. oxygen inhibited) acidic, hydrophilic adhesive layer (Rueggeberg & Margeson 1990; Suh 2004).

In this present study, two three-step total-etch dentin adhesives were tested (Table 1). Thus, the adhesive layers for both dentin adhesives were basically hydrophobic. However, All-Bond 2 bonding resin (D/E Resin) contain 30% 2-hydroxyethyl methacrylate (HEMA), which is a small monomer that is in widespread use, not only in dentistry

(Van Landuyt *et al.* 2007). Its popularity in medical applications must be attributed to its relatively good biocompatibility, even though the uncured monomer is notorious for its high allergenic potential. Uncured HEMA presents as a fluid that is well solvable in water, ethanol and/or acetone. Moreover, HEMA has been described to be able to evaporate from the adhesive solutions, though only in very small amounts (Pashley *et al.* 1998). Another important characteristic of HEMA is its hydrophilicity. Even though this monomer cannot be used as a demineralizing agent, its hydrophilicity makes it an excellent adhesion-promoting monomer. By enhancing wetting of dentin, HEMA significantly improves bond strengths. Nevertheless, both in uncured and cured state, HEMA will readily absorb water. HEMA in the uncured adhesive may absorb water, which can lead to dilution of the monomers to the extent that polymerization is inhibited (Van Landuyt *et al.* 2007). HEMA fixed in a polymer chain after polymerizing will still exhibit hydrophilic properties and will lead to water uptake with consequent swelling and discoloration (Burrow *et al.* 1999). Apart from the water uptake, which adversely influences the mechanical strength, high amounts of HEMA will result in flexible polymers with inferior qualities(Van Landuyt *et al.* 2007).

Therefore, it seems that the hydrophilicity of the All-Bond 2 adhesive layer affected the microtensile bond strength values, depending on the thickness of the adhesive layer (Tay & Pashley 2003). This finding suggests that the thickness of 50 μm did not create a high-quality adhesive layer. On the other hand, the thickness of 200 μm could increase the degree of hydrophilicity of the bonding interface. According the manufacturer, the All-Bond 3 bonding resin, which does not contain HEMA, is more hydrophobic than that of All-Bond 2. Hence, the influence of the adhesive layer thickness of the material was minimal.

Although one reason for difference in the microtensile bond strength in All-Bond 2 may be difference in the monomer conversion of the adhesive layers (Yan *et al.* 2010), polyHEMA is basically a flexible porous polymer even after polymerization (Van Landuyt *et al.* 2007). As such, high concentrations of HEMA in an adhesive may have deteriorating effects on the mechanical properties of the resulting polymer. HEMA also lowers the vapor pressure of water, and probably also of alcohol. High amounts may therefore hinder good solvent evaporation from adhesive solutions (Pashley *et al.* 1998).

Like all methacrylates, HEMA is vulnerable to hydrolysis, especially at basic pH, but also at acidic pH. HEMA is very frequently added to adhesives, not only to ensure good wetting, but also because of its solvent-like nature. This property improves the stability of solutions containing hydrophobic and hydrophilic components and will keep ingredients into solution (Van Landuyt *et al.* 2005). Although, the incorporation of HEMA into dentin adhesives is sometimes indispensable, the amount should be carefully controlled not to make the materials too hydrophilic (Tay & Pashley 2003). According to a previous study (Kim *et al.* 2014), dentin surface moisture has a crucial effect on the bond strength of resin materials. This issue was not included in this study; further studies are needed.

IV. CONCLUSION

The influence of adhesive layer thickness on microtensile bond strength to dentin was found to be material-specific. As for All-Bond 2, the relatively thick adhesive layer produced an enhanced resin bonding to dentin compared with the thin adhesive layer. For All-Bond 3, the thickness of the adhesive layer did not significantly affect the microtensile bond strength. These results seem to have relation with the incorporation of the hydrophilic monomer HEMA into the bonding resins.

V. REFERENCES

Burrow MF, Inokoshi S, Tagami J (1999). Water sorption of several bonding resins. Am J Dent 12:295-298.

Cadenaro M, Breschi L, Antoniolli F *et al.* (2008). Degree of conversion of resin blends in relation to ethanol content and hydrophilicity. Dent Mater 24:1194-1200.

Di Hipólito V, Rodrigues FP, Piveta FB *et al.* (2012). Effectiveness of self-adhesive luting cements in bonding to chlorhexidine-treated dentin. Dent Mater 28:495-501.

El Zohairy AA, De Gee AJ, Mohsen MM, Feilzer AJ (2005). Effect of conditioning time of self-etching primers on dentin bond strength of three adhesive resin cements. Dent Mater 21:83-93.

Hiraishi N, Yiu CK, King NM, Tay FR (2009). Effect of pulpal pressure on the microtensile bond strength of luting resin cements to human dentin. Dent Mater 25: 58-66.

Kim DH, Son JS, Jeong SH, Kim YK, Kim KH, Kwon TY (2015). Efficacy of various cleaning solutions on saliva-contaminated zirconia for improved resin bonding. J Adv Prosthodont 75:85-92.

Kim YK, Min BK, Son JS, Kim KH, Kwon TY (2014). Influence of different drying methods on microtensile

bond strength of self-adhesive resin cements to dentin. Acta Odontol Scand 72:954-962.

Kwon TY, Bagheri R, Kim YK, Kim KH, Burrow MF (2012). Cure mechanisms in materials for use in esthetic dentistry. J Investig Clin Dent 3:3-16.

Mazzitelli C, Monticelli F, Osorio R, Casucci A, Toledano M, Ferrari M (2008). Effect of simulated pulpal pressure on self-adhesive cements bonding to dentin. Dent Mater 24:1156-1163.

Pashley EL, Zhang Y, Lockwood PE, Rueggeberg FA, Pashley DH (1998). Effects of HEMA on water evaporation from water-HEMA mixtures. Dent Mater 14: 6-10.

Rueggeberg FA, Margeson DH (1990). The effect of oxygen inhibition on an unfilled/filled composite system. J Dent Res 69:1652-1658.

Suh BI (2004). Oxygen-inhibited layer in adhesion dentistry. J Esthet Restor Dent 16:316-323.

Suh BI, Feng L, Pashley DH, Tay FR (2003). 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 5:267-282.

Tay FR, Pashley DH (2003). Have dentin adhesives become too hydrophilic? J Can Dent Assoc 69:726-731.

Tay FR, Suh BI, Pashley DH, Prati C, Chuang SF, Li F (2003). 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 5:91-105.

Van Landuyt KL, De Munck J, Snauwaert J et al. (2005). Monomer-solvent phase separation in one-step self-etch adhesives. J Dent Res 84:183-188.

Van Landuyt KL, Snauwaert J, De Munck J et al. (2007). Systematic review of the chemical composition of contemporary dental adhesives. Biomaterials 28:3757-3785.

Yan YL, Kim YK, Kim KH, Kwon TY (2010). Changes in degree of conversion and microhardness of dental resin cements. Oper Dent 35:203-210.

Yip HK, Tay FR, Ngo HC, Smales RJ, Pashley DH (2001). Bonding of contemporary glass ionomer cements to dentin. Dent Mater 17:456-470.