The influence of moisture control on bond strength of composite resin treated with self-etching adhesive system

Myoung Uk Jin, Young Kyung Kim, Jeong-won Park
Department of Conservative Dentistry, School of Dentistry, Kyungpook National University

I. Introduction

A goal of restorative dentistry is to develop adhesive restorative materials that are durable and provide an effective seal at the restoration/tooth interface. To achieve this purpose, many enamel-dentin bonding systems were introduced and improved in a new formula. These new adhesive materials not only attempt to improve the quality of the bond, but also simplify the clinical procedures.

Most recent innovative changes of bonding concepts are the "total-etch" and "wet-bonding" techniques, and these methods have been used for many years. However, when using the wet bonding technique it is difficult to maintain the opti-
mal level of moisture.

In order to simplify the clinical procedures and improve the quality of the bonding, a self-etching adhesive system was developed. It was developed to eliminate the conditioning, rinsing, and drying steps that may prove to be critical and difficult to standardize in operative conditions. Also it has been reported that the self-etching primer system produce high bond strengths to dentin, independent of regional differences—deep vs superficial dentin, crown vs root dentin.

Theoretically, the new acidic part of the primer dissolves the smear layer, incorporating it into the mixture, as it demineralize the dentin and encapsulates the collagen fibers and hydroxyapatite crystals. The acidity of the primer is less than that of 32% or 37% phosphoric acid gels, but is sufficient to etch through smear layers into the underlying enamel or dentin. Barkmeier et al. examined the effect of this system, an acidic primer, on enamel and dentin through shear bond strength testing and evaluation of marginal microleakage. SEM examination revealed resin penetration into both enamel and dentin surfaces, indicating that adequate conditioning was achieved and produced high bond strengths to both enamel and dentin. Since the etching and priming processes run parallel to each other, depth of demineralization and penetration depth of the bonding agent are identical, and it should minimize voids, as all the entire region would be completely encapsulated by the resin primer. As a result, light curing of these penetrated monomers and copolymerization of the overlying resin bonding agent and composite resin form a continuous bond with the tooth surface which is capable of resisting the effect of microleakage.

But, in clinical usage of self-etching primer system, there are some problems that must be solved. First, the enamel etching, using the self-etching primer system, is controversial. The acidic part of the primer is neutralized by calcium and phosphate ions released during demineralization. When the water is evaporated during air drying, the concentrations of solubilized calcium and phosphate within the primer may exceed the solubility product constants for a number of calcium and phosphate. Thus it is conceivable that residues of the primer or possibly precipitates of calcium phosphates could remain on the tooth surface and thereby masked the etching pattern. Therefore, demineralization is self-limiting, in that the high concentration of these ions tends to inhibit further dissolution of hydroxyapatite.

Second, manufacturer recommended dry bonding technique, to avoid diluting the acidic component of the primer with excess surface water. It has already been proven that proper application of the adhesive primer plays an important role in achieving good bond strength. However, clinicians have been using the wet bonding technique for many years, so they are still becoming adept at using this techniques. Sometimes, adequate moisture control is difficult or not possible in some clinical situations. In these condition, the tooth surface is moist and the bonding quality can be affected. But little research has been conducted regarding the effects of moisture when using the self-etching adhesive system.

The purpose of this study is to determine the influence of moisture control on bond strength of the self-etching adhesive system by measuring shear bond strengths of composite resin to enamel and dentin treated with self-etching adhesive system.

II. Materials and Methods

Ninety-six extracted, noncarious human molars stored at 4°C in isotonic saline were used in this study. The ages of the patients were not known and the reasons for extraction ranged from periodontally compromised teeth to impacted teeth. During the last 24 hours before beginning the experiment, they were kept in distilled water.

The teeth were embedded in auto-polymerizing acrylic resin (Orthodontic Resin, Dentsply/Detray, Konstanz, Germany) molds so that the prepared enamel and dentin surfaces were 2 mm above the acrylic resin cylinders, and placed in tap water to reduce the temperature rise from the exothermic polymerization reaction.
After the resin had completely polymerized, the occlusal surfaces of the teeth were ground perpendicular to the long axis of the tooth on a water-cooled, model trimming wheel to create flat enamel and dentin surfaces. Then the enamel and dentin surfaces were hand finished with using wet 600-grit silicon carbide abrasive papers using twenty 15 cm long strokes. After ultrasonic cleaning with distilled water for 3 minutes to remove the excess debris, these surfaces were washed and dried with oil-free compressed air (Hotman, Dentro, Tokyo, Japan). The teeth were randomly divided into eight groups of 12 teeth each and treated in the manner (Table 1). Then the Clearfil SE Bond primer (Kuraray Co., Ltd., Osaka, Japan) (Table 2) was applied to the tooth surface with Microbrush (Int’l, Co., Ltd., Dungarvan, Waterford, Ireland) and allowed to sit for 20 seconds according to the manufacturer’s recommendations.

After primed tooth surface was dried with oil-free compressed air for 5 seconds from a distance of 10 cm, the bonding agent was applied to the surface and irradiated with curing unit (Spectrum 800, Dentsply/Detray, Konstanz, Germany) for 10 seconds, with the intensity set at 400 mW/cm².

A mount jig (Ultradent Product Inc., South Jordan, Utah, U.S.A.) with an internal ring of 2.3798 mm in diameter and height of 2.0 mm was placed against the tooth surface and stabilized with an alignment tube. A resin composite (Clearfil AP-X, Kuraray Co., Ltd., Osaka, Japan) was packed into the mold and light-cured for 40 seconds. After polymerization, the alignment tube and mold were removed and the specimens were placed in 37°C distilled water. Twenty-four hours after storage, the specimens in each group were tested in shear mode using a chisel-shaped rod in an Instron testing machine (Type 4411, Instron Corp., Canton, Massachusetts, U.S.A) at a crosshead speed of 1 mm/minute.

The data for each group were subjected to one-

### Table 1. Experimental groups following drying condition of enamel and dentin surfaces

| Group   | Enamel / Dentin drying condition                                      |
|---------|-----------------------------------------------------------------------|
| Group 1 | dried for 30 min. at room temperature dry no visible moisture was seen |
| Group 2 | dried for 5s with oil-free air dry from a distance of 10 cm, no visible moisture, excessive desiccation avoided |
| Group 3 | dried for 1s with oil-free air dry from a distance of 10 cm, removed the excessive moisture |
| Group 4 | blot dry with cotton pellet, whole surface was visibly moist          |

### Table 2. Composition of Clearfil SE Bond

| Clearfil SE Bond |
|------------------|
| PRIMER | BOND |
| 10-Methacryloxydeyl dihydrogen phosphate (MDP) | 10-Methacryloxydeyl dihydrogen phosphate (MDP) |
| 2-Hydroxyethyl methacrylate (HEMA) | Bis-phenol A diglycidylmethacrylate (Bis-GMA) |
| Hydrophilic dimethacrylate | 2-Hydroxyethyl methacrylate (HEMA) |
| dl-Camphorquinone | dl-Camphorquinone |
| N,N-diethanol-p-toluidine | Hydrophobic dimethacrylate |
| Water | N,N-diethanol-p-toluidine |
| silanated colloidal silica |
way ANOVAs followed by the Duncan’s multiple range test at p<0.05 to make comparisons among the groups.

### III. Results

The results of the shear bond strength tests to enamel are shown in Table 3, and those of dentin in Table 4.

In both the enamel and dentin experimental groups, there were statistically significant differences (p<0.05) between these groups which were dried for 30 minutes at room temperature (E1, D1) or for 5 seconds with oil-free air (E2, D2) and those which were dried for 1 second with oil-free air (E3, D3) or blotted dry (E4, D4) (p<0.05).

### Table 3. Shear bond strength of resin composite to enamel treated with Clearfil SE Bond (MPa) (Mean S.D.)

| Group         | n  | Shear bond strength |
|---------------|----|---------------------|
| E1 (30min, dry) | 12 | 27.5±5.5            |
| E2 (5s, air dry) | 12 | 26.2±6.3            |
| E3 (1s, air dry) | 12 | 18.6±4.1*           |
| E4 (blot dry)   | 12 | 17.3±4.3*           |

*statistically significant difference by One-way ANOVA (p<0.05).

### Table 4. Shear bond strength of resin composite to dentin treated with Clearfil SE Bond (MPa) (Mean S.D.)

| Group         | n  | Shear bond strength |
|---------------|----|---------------------|
| D1 (30min, dry) | 12 | 24.8±4.6            |
| D2 (5s, air dry) | 12 | 25.7±3.2            |
| D3 (1s, air dry) | 12 | 17.1±3.1*           |
| D4 (blot dry)   | 12 | 16.8±2.4*           |

*statistically significant difference by One-way ANOVA (p<0.05).

### IV. Discussion

Recently developed adhesive systems are characterized by their simple application procedure. Among simplified adhesives the self-etching approach currently appears most promising in avoiding inadequate hybridization due to collagen collapse in over-dry conditions or due to residual solvent in over-wet conditions. In self-etching adhesive system, simplification of the clinical application procedure is obtained not only by reduction of application steps, but by omission of postconditioning rinsing phase. As a rinse phase is not required, the risk of substrate contamination with saliva or other dentin-porosity blocking agents is virtually eliminated. As a supplementary advantage, the controversy of post-conditioning drying or keeping the dentin moist, as in wet-bonding technique is avoided. The actual rationale behind these systems is to superficially demineralize dentin and to simultaneously penetrate it to the depth of demineralization with monomers that can be polymerized in situ. However, in clinical situation, the complete dry of cavity is difficult.

Based on their interaction with dentin, there are two primary types of self-etching adhesive system distinguished.

The first group of moderate self-etching systems is constituted by Clearfil SE Bond, Clearfil Liner Bond 2V, F2000 primer/adhesive systems (3M dental products Inc., St. Paul, MN, U.S.A), and Unifil Bond (GC, Tokyo, Japan), all of which have a pH of about 2. The latest self-etching
primer system, Clearfil SE Bond (Kuraray, Osaka, Japan, known in Japan as Clearfil Mega Bond) combines the two separate primer solutions of Clearfil Liner Bond 2V into a single bottle. This reduces the etching time from 30s to 20s.

The second group consists of adhesive systems such as Prompt L-Pop (ESPE, Seefeld, Germany), a self-etching adhesive, and Prime & Bond NT (Dentsply/DeTrey, Konstanz, Germany) in which a nonrinse conditioner (NRC) is used as a self-etching primer treatment. These adhesives have a pH of 1 or less and interact more profoundly with dentin. Despite the fact that these “strong” self-etching primers are not rinsed off, their interfacial ultramorphologic features closely resemble those of total-etch systems that use phosphoric acid that is rinsed off. Based on this fact, it might be assumed that water dilution of acidic composition in Clearfil SE Bond would influence the etching ability. But there has not been enough research conducted in water dilution and etching ability to determine this fact.

This study evaluated the shear bond strength of self-etching adhesive system, Clearfil SE Bond, in the presence of varying amount of water that are to be expected during clinical bonding procedures.

According to the manufacturer’s recommendations, dry bonding is an effective technique to use with the Clearfil SE Bond system. But, the degree of dryness is not stipulated in the manufacturer’s recommendation. Following the results of this experiment, both enamel and dentin, 5 seconds and 30 minutes dry groups showed higher bond strength than 1 second and blot dry groups. 1 second air drying and blot drying with a cotton pellet are both representative of procedures which are used in the wet bonding technique. And it was found that 5 seconds of air drying was sufficient time for the extra moisture to evaporate from the tooth surface. As expected, moisture had a detrimental effect on the bond strength of the self-etching primer.

The low bond strengths of the blot drying and 1s air drying groups might have been caused by the resulting dilution and incomplete diffusion of primer components. Such dilution and incomplete diffusion could preclude the production of high-quality polymers, thus compromising bond strength.

The high moisture condition can adverse effect on Clearfil SE system. Camphoroquinone (CQ) is widely used as a photoinitiator for bonding systems and it is also used in Clearfil system. It is impossible for CQ to initiate the polymerization of water-soluble monomers in the water. Furthermore it is doubtful that CQ can sufficiently initiate the polymerization of diffused monomer, inside the dentin due to its water content and the fact that it has a lower partition coefficient than resin. Assuming that water may be retained in the HEMA, and that the diffusion of the light-activating components may be delayed by interference with the water, it seems reasonable to suspect that large variations in cure may exist within the hybrid layer. To solve this problem, 2-hydroxy-3-(3,4-dimethyl-9-oxo-9H-thioxanthen-2-yloxy)-N,N,N-Trimethyl-1-1-propanaminium chloride (QTX) was introduced as an alternative to CQ. It is supposed that QTX could initiate sufficient polymerization of diffused monomers inside the dentin despite the presence of water and can improve the polymerization degree of the diffused monomers, result in improve bond strengths to dentin. Of course, there is not sufficient research in this area to come to a definite conclusion.

Clearfil SE Bond system contains water as a primer component. When water is used as a solvent, the HEMA molecules do not saturate the collagen mesh as quickly as it would if an acetone-HEMA solvent were used. Instead, the water and HEMA molecules compete for space within the collagen mesh. The presence of water inside the collagen mesh may cause a lower degree of polymerization than occurs with the HEMA/Bis-GMA bonding resin. This results in a lower molecular weight of the poly-HEMA and a weaker interpenetrating network. If excessive moisture was present on the primed tooth surface, Bis-GMA component of bonding mixes poorly with water. Therefore, the Bis-GMA monomer forms an emulsion with the water, and during
curing process, a Bis-GMA “sponge” or Bis-GMA droplets cure depending on the geometrical shape of the Bis-GMA phase. Thus, an increase in water will increase the pore size of the “sponge” or increase the distance between the droplets. Therefore, the reduced conversion caused by water contamination also reduces bond strength17).

Jacobson et al17) tested the effects of water on the polymerization of a mixture of adhesive monomers by measuring the conversion rate of double bonds by FT-IR. In the presence of no water, the conversion rate was 53.5%. Although addition of 5% water only reduced the conversion rate to 52.6%, 10% water reduced it to 31.5%, and 20% water further reduced it to 22.7%. Thus, it appears that excess residual water may adversely affect the polymerization of the resin, thereby lowering its final mechanical properties.

According to Pashley et al18), the rate of evaporation of water from water–HEMA mixtures is inversely related to the relative humidity of the environment and to the vapor pressure of water. Relative high humidity lowers the net flux of water vapor, so residual water might interfere with polymerization of adhesive monomers, thereby lowering the quality of the hybrid layer. The degree of environmental humidity is high and comparable to that of the oral cavity when no rubber dam is used, whereas with rubber dam use the environmental humidity will be similar to that of the ambient air in the operatory20). Thus, rubber dam application while using the bonding system is a very important procedure.

MDP is incorporated in both primer and bonding agents of the Clearfil SE Bond system (Table 2). A similar phenomenon has been reported as “MDP balls” when an aqueous mixture of 10-methacryloyloxydecyl dihydrogen phosphate (MDP) and 30% HEMA was applied to smear layer retained dentin20) and the same such micelles resulted. Such interface deficiencies undoubtedly weaken the resin–dentin bond and result in incompletely sealed tubules.

Excessive water that was not adequately removed during priming appeared to cause deterioration of the bonded assembly along the hybrid layer–resin interface21), as well as phase separation of the hydrophobic and hydrophilic monomer components, which caused blistering and the formation of the globules at the resin–dentin interface9).

In treating more complex cavities like cervical abrasion or gingival box-like cavities, moisture control of operative site is difficult to perform. A simple, flat bonding site used in this study does not exist in the clinical situations. Therefore, using an air syringe to remove excess surface water from complex cavities could produce areas of ideal surface moisture, but could also result in over-wet or over-dry line angles11). Therefore, nonuniform surface condition might produced, leading to lower bond strengths. So, when air drying before the application of self-etching system, warm and oil-free air is preferred, careful usage, sufficient drying time, and visual examination are recommended.

The self-etching primer systems efficiently bonded to dry tooth surfaces. Thus before any bonding procedure is begun, adequate isolation and moisture control of the substrate to be bonded to must be achieved. Following the result of this experiment, using an oil-free compressed air for 5s sufficiently dry the tooth surface without causing excessive drying the tooth structure. In practical view, clinicians should be cautious not to overwetting the dentin surface, such as puddling around the line angles of cavity. More research is needed to examine the varying drying methods and times for better bonding strengths when using the self-etching adhesive system.

V. Conclusion

Recently, self-etching adhesive systems have been developed and bonding procedures simplified into two steps, which are simultaneously applied to both enamel and dentin. These systems are easy to use and have the potential for good clinical success. It has been reported that the self-etching adhesive systems produce high bond strength to enamel and dentin.

The purpose of this study was to study the
influence of moisture control on bond strength of self-etching primer by measuring enamel and dentin shear bond strengths.

96 human molars were divided into 8 groups. Group E1 (Enamel, 30min room temperature dry), Group E2 (Enamel, 5s air dry), Group E3 (Enamel, 1s air dry), Group E4 (Enamel, blot dry), Group D1 (Dentin, 30min room temperature dry), Group D2 (Dentin, 5s air dry), Group D3 (Dentin, 1s air dry), Group D4 (Dentin, blot dry).

Clearfil SE Bond primer was applied to the enamel and dentin surfaces and after 20s surfaces were air dried. Then the bonding agent was applied and light cured for 10 seconds. A resin composite (Clearfil AP-X, Kuraray Co., Ltd., Osaka, Japan) was packed into the mold and light-cured for 40 seconds. Twenty-four hours after storage, the specimens were tested in a shear bond test.

Both in enamel and dentin, 30 min drying and 5 seconds air drying groups showed significantly higher bond strengths than 1s air drying and blot drying groups (p<0.05). Using the self-etching adhesive system, this study concluded that the complete dry bonding technique is effective on both enamel and dentin. When the excess moisture is existing on the bonding surface, shear bond strength decreased rapidly. So in clinical application of self-etching adhesive system, the cautious moisture control is needed.

References

1. Pashley, D., and Sano, H.: Adhesion testing of dentin bonding agents. Dent. Mater., 11:117-125, 1995.
2. Yoshikawa, T., Sano, H., Burrow, M., Tagami, J., and Pashley, D.: Effects of dentin depth and cavity configuration on bond strength. J. Dent. Res., 78: 898-905, 1999.
3. Tay, F., Carvalho, R. and Sano, H.: Effect of smear layers on the bonding of a self-etching primer to dentin. J. Adhes. Dent., 2: 99-116, 2000.
4. Tay, F., Sano, H., Carvalho, R., Pashley, E., and Pashley, D.: An ultrastructural study of the influence of acidity of self-etching primers and smear layer thickness on bonding to intact dentin. J. Adhes. Dent., 2: 83-98, 2000.
5. Barkmeier, W., Los, S., and Triolo, P.: Bond strength and SEM evaluation of Clearfil Liner Bond 2. Am. J. Dent., 8: 289-293, 1995.
6. Hannig, M., Reinhardt, K., and Bott, B.: Self-etching primer vs phosphoric acid: An alternative concept for composite-to-enamel bonding. Oper. Dent., 24: 172-180, 1999.
7. Hayakawa, T., Kikuteke, K., and Nemoto, K.: Influence of moisture control on bond strength of composite resin to polished dentin and enamel. Dent. Mater., 14: 99-105, 1998.
8. Samir, E., Bishara, S., and von Wald, L.: Effect of a self-etch primer/adhesive on the shear bond strength of orthodontic brackets. Am. J. Orthod. Dentofacial Orthop., 119: 621-624, 2001.
9. Ferrari, M., Mannocci, F., Vichi, A., and Davidson, C.: Effect of two etching times on the sealing ability of Clearfil Liner Bond 2 in Class V restorations. Am. J. Dent., 10: 66-70, 1997.
10. Pashley, E., Zhang, Y., Lockwood, P., Rueggeberg, F., and Pashley, D.: Effects of HEMA on water evaporation from water-HEMA mixtures. Dent. Mater., 14: 6-10, 1998.
11. Tay, F., Gwinnett, J., and Wei, S.: Micromorphological spectrum from overdrying to overwetting acid-conditioned dentin in water-free, acetone-based, single-bottle primer/adhesives. Dent. Mater., 12: 236-244, 1996.
12. Nakajima, M., Ogata, M., Harada, N., Tagami, J., and Pashley, D.: Bond strength of self-etching primer adhesive to in vitro - demineralized dentin following mineralizing treatment. J. Adhes. Dent., 2: 29-38, 2000.
13. Miller, R.: Laboratory and clinical evaluation of a self-etching primer. J. Clin. Ortho., 42-45, 2001.
14. Ferrari, M., Mannocci, F., Kugel, G., and Garcia-Godoy, F.: Standardized microscopical evaluation of the bonding mechanism of NRC/Prime & Bond NT. Am. J. Dent., 12: 77-83, 1999.
15. Schwartz, R., Summitt, J., and Robbins, J.: Fundamentals of Operative Dentistry: 2nd ed. Quintessence Publishing Co, Inc., Illinois (2000) pp.178-221.
16. Hayakawa, T., Kikuteke, K., and Nemoto, K.: Effectiveness of the addition of water-soluble photoinitiator into the self-etching primers on the adhesion of a resin composite to polished dentin and enamel. Dent. Mater. J., 18: 324-333, 1999.
17. Jacobsen, T., and Soderholm, K.: Some effects of water on dentin bonding. Dent. Mater., 11:132-136, 1995.
18. Yukitani, W., Chigira, H., and Hasegawa, T.: Self-etching dentin primers containing Phenyl-P. J. Dent. Res., 73:1088-1095, 1994.
19. Koibuchi, H., Yasuda, N., and Nakabayashi, N.: Bonding to dentin with a self-etching primer: the effect of smear layers. Dent. Mater., 17: 122-126, 2001.
20. Tay, F., Gwinnett, J., and Wei, S.: The overwet phenomenon: A scanning electron microscopic study of surface moisture in the acid-conditioned, resin-dentin interface. Am. J. Dent., 9: 109-1141, 1996.
21. Paul, S.: Effect of water content on the physical properties of model dentine primer and bonding resins. J. Dent., 27: 209-214, 1999.