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Original

Effect of thermal cycling on bond strengths of single-step self-etch adhesives to bovine dentin

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Abstract: The purpose of this study was to clarify the effect of thermal cycling on dentin bond strengths of single-step self-etch adhesives. Five commercially available single-step self-etch systems were used. The adhesives were applied to the dentin surfaces of bovine incisors, and then light-irradiated. Resin composites were condensed into a mold and light-irradiated. Bonded specimens were divided into two groups and stored in water at 37˚C for 24 h without thermal cycling, or in water at 37˚C for 24 h followed by 10,000 thermal cycles between 5˚C and 60˚C. Ten samples per group were tested for shear strength at a crosshead speed of 1.0 mm/min. The data were analyzed by Student's *t* test and Tukey HSD test at a probability level of 0.05. After 24 h of storage in water, the mean dentin bond strengths ranged from 9.3 MPa to 14.0 MPa. After 10,000 thermal cycles, the mean bond strengths remained unchanged. Failure after the test was commonly due to adhesive breakdown associated with partial cohesive failure of the resin. The present results suggest that the benefit of using single-step self-etch systems, in terms of simplifying the clinical procedure, might be acceptable even after thermal stresses. (J. Oral Sci. 48, 63-69, 2006)

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Introduction

Conventional dentin bonding systems require sequential application of conditioner, dentin primer and bonding agent in several clinical steps. To reduce technique-sensitive and materials-related factors that affect bond strength, a self-etch approach involving either one or two-step application has been developed (1). These simplified systems aim to reduce technique-sensitivity by reducing the number of clinical steps involved. Self-etch adhesives are applied to the tooth surface prior to resin composite placement, to ensure maximum adhesion by improving monomer penetration into the hydrophilic dentin substrate, and to improve wettability of the tooth surface by the resin components (2). Single-step self-etch adhesive systems form a continuous layer by demineralization of the superficial dentin substrate, followed by resin monomer penetration into the etched dentin (3).

Previously, the application of single-step self-etch adhesives to dentin resulted in retention of a smear layer and insufficient bond strength (4). By contrast, higher bond strengths were obtained for two-step self-etch primer adhesive systems through the introduction of a submicron resin tag (5). This raises the question of whether the creation of a dentin/adhesive interaction might be sufficient to create stable adhesion after thermal cycle stress (6). The evaluation of bonding durability is important, as the bond

between the restorative material and the tooth substrate has a significant impact on the clinical success of a restoration (7). Long-term storage of a bonded specimen in water or subjecting it to thermal cycling can give some insight into the temperature-dependent degradation of the material (8). The thermal cycling test involves subjecting specimens to extreme temperatures that simulate intra-oral conditions to induce stress between a tooth substrate and a restorative material by generating contraction stress (9). The effect of thermal cycling on the bond strength of multi-step bonding systems reportedly depends on the bonding system used and the number of thermal cycles (10-12).

The purpose of the present study was to determine the effect of thermal cycling on bond strengths of single-step self-etch adhesive systems to bovine dentin, by measuring the shear bond strength, and through scanning electron microscopy (SEM) observations of the fractured surface after bond strength measurement.

Materials and Methods

The following single-application self-etch adhesive systems, with a combination of resin composites, were used: Adper Prompt L-Pop/ Filtek Z250 (3M ESPE, St. Paul, MN, USA), AQ Bond Plus/ Metafil C (Sun Medical, Shiga, Japan), Fluoro Bond Shake One/Beautifil (Shofu Inc., Kyoto, Japan), G-Bond/Gradia Direct (GC Corp., Tokyo, Japan), and One-Up Bond F Plus/Palfique Estelite ∑ (Tokuyama Dental, Tokyo, Japan), as shown in Table 1. All adhesive systems were used in combination with the relevant manufacturer's restorative resin. The application protocols suggested by each manufacturer are listed in Table 2.

A visible-light-activating unit, Optilux 501 (SDS Kerr, Danbry, CT, USA), was used. The power density (800 mW/cm^2) of the curing light was checked with a dental radiometer (Model 100, SDS Kerr) before manufacturing the specimens.

Mandibular incisors that had been extracted from 2- to 3-year-old cattle, and stored frozen for up to 2 weeks, were used as a substitute for human teeth (13, 14). After removing the roots with a slow-speed saw and a diamond-impregnated disk (Isomet, Buehler Ltd., Lake Bluff, IL, USA), the pulps were removed, and the pulp chamber of each tooth was filled with cotton to avoid penetration of the embedding media. The labial surfaces of the bovine incisors were ground on wet 240-grit SiC paper to achieve a flat dentin surface. Each tooth was then mounted in cold-curing acrylic resin (Tray Resin II, Shofu Inc., Kyoto, Japan) to expose the flattened area, and submerged in tap water to reduce any temperature rise caused by the exothermic polymerization reaction of the acrylic resin. The final finish was accomplished by grinding the teeth on wet 600-grit SiC paper. After ultrasonic cleaning with distilled water for 1 min to remove any excess debris, the surfaces were washed and dried with oil-free compressed air.

A piece of double-sided adhesive tape, bearing a hole 4 mm in diameter, was firmly attached in order to define the designated bonding area. The adhesive was applied to the dentin surface according to the instructions of each manufacturer (Table 2). After the adhesive had been applied, the surfaces were dried with oil-free compressed air, which was delivered at a pressure of 0.2 MPa from 5 cm above the dentin surface for 5 sec using a three-way syringe. The surfaces were then irradiated using a curing unit. A Teflon (Sanplatec Corp., Osaka, Japan) mold (2.0 mm in height and 4.0 mm in diameter) was used to form and then hold the restorative resin on the dentin surface. The resin composite was condensed into the mold and cured for 30 sec. The Teflon mold and adhesive tape were removed from the specimens 10 min after light-irradiation. The bonded specimens from each set of materials were divided into two treatment groups (n = 10 in each) as follows: group 1 specimens were stored in water at 37˚C for 24 h after placement, without thermal cycling; and group 2 specimens were stored in water at 37˚C for 24 h, followed by 10,000 thermal cycles between 5˚C and 60˚C. The dwelling time in the water bath was 30 sec and the transfer time was 5 sec.

After treatment, the specimens in each group were tested for shear strength using a knife-edge testing apparatus and a universal testing machine (Type 4204, Instron Corp., Canton, MA, USA) at a crosshead speed of 1.0 mm/min. The shear bond-strength values (MPa) were calculated from the peak load at failure divided by the specimen surface area.

After testing, the specimens were examined using an optical microscope (SZH-131, Olympus Ltd., Tokyo, Japan) at ×10 magnification in order to identify the location of the bond failure. The type of bond failure was determined on the basis of the percentage of substrate-free material, as follows: adhesive failure, cohesive failure of the composite, cohesive failure of the adhesive resin, or cohesive failure of the dentin (15).

The results were analyzed by calculating the mean shear bond strength (MPa) and standard deviation for each group. The data for each group were subjected to Student's *t* test and Tukey's HSD test at a probability level of 0.05. All statistical analyses were carried out using the Sigma Stat® Ver. 3.1 (SPSS Inc., Chicago, IL, USA) software system.

The ultrastructure of the dentin surfaces was examined by field-emission (FE)-SEM. Briefly, fractured specimens

were dehydrated after the bond strength measurements in ascending concentrations of *tert*-butanol (50% for 20 min, 75% for 20 min, 95% for 20 min, and 100% for 2 h), and then transferred to a critical-point dryer. The surfaces were coated in a vacuum evaporator (Quick Coater Type SC-701, Sanyu Denshi Inc., Tokyo, Japan) with a thin film of gold. The specimens were then observed under FE-SEM (ERA-8800FE, Elionix Ltd., Tokyo, Japan).

Table 1 Single-step self-etch adhesive systems

Adhesive (Manufacturer)	Main components	Lot No.
Adper Prompt L-Pop (3M ESPE)	Methacrylated phosphoric esters, Bis-GMA,CQ, initiator, stabilizer, 2-HEMA, polyalkenoic acid, water	127613
AQ Bond Plus (Sun Medical)	Water, acetone, 4-META, UDMA, HEMA, MMA, initiator *p*-toluenesulfinic acid sodium salt	FW1 FX1
Fluoro Bond Shake One (Shofu Inc.)	Pre-reacted glass-ionomer filler, fluoroaluminosilicate glass, 4-AET, 4-AETA, bis-GMA initiator, water, solvent,	A: 551F-2 B: 551F
G-BOND (GC Corp.)	4-MET, UDMA, acetone, water silanated colloidal silica, initiator	0403191
One-Up Bond F Plus (Tokuyama Dental)	Water, MAC-10, HEMA, MMA, multifunctional methacrylic monomer fluoroaluminosilicate glass, photo initiator, aryl borate catalyst	A: 004 B: 504

Table 2 Application protocols of single-step self-etch adhesive systems

Adhesive system	Application protocol
Adper Prompt L-Pop (Blister-packed)	Activate blister pack by emptying the liquid out of the red blister into the yellow blister. The activated solution was applied to dentin for 15 sec with moderate finger pressure. Gentle stream of air to dry and apply second coat of adhesive. Gently air dry and light irradiation for 10 sec.
AQ Bond Plus (Single bottle with sponge)	Dispense one drop of liquid into well containing one piece of sponge. Apply the mixed sponge to dentine for 20 sec. Gently air dry for 5~10 sec, relatively strong air dry for 5~10 sec, and light irradiation for 10 sec.
Fluoro Bond Shake One (Two bottles)	Mix equal amounts of bond agent A and B. Apply to dentin for 20 sec. Briefly air dry and light irradiation for 10 sec.
G-Bond (Single bottle)	Apply sufficient amount of adhesive for 10 sec. Strong air dry and light irradiation for 10 sec.
One-Up Bond F Plus (Two bottles)	Mix equal amounts of the bond agents A and B for until a pink homogenous liquid mixture was obtained. Apply to dentin for10 sec with agitation and light irradiation for 10 sec.

Results

The mean shear bond strengths to bovine dentin, and the causes of failure after the test, are shown in Table 3. After 24 h storage in water, the mean dentin bond strengths of the single-step self-etch adhesive systems ranged from 9.3 MPa to 14.0 MPa. The Fluoro Bond Shake One showed the numerically highest bond strength. No changes in bond strength after 10,000 thermal cycles were observed for the adhesive systems employed (from 9.6 to 14.3 MPa).

The predominant mode of failure was adhesive breakdown at the dentin surface associated with partial cohesive failure of the resin, except for Fluoro Bond Shake-One which exhibited partially cohesive failure in dentin. The FE-SEM observations of the fractured resin surfaces (Fig. 1) revealed adhesive failure between the dentin and the adhesive. In addition, several small cracks were observed on the cured adhesive resin at higher magnification (Fig. 2).

Discussion

It has been reported that the dentin bond strength of third generation adhesive systems decreases as the number of thermal cycles increases (16); this could be attributable to the presence of a region of demineralized dentin that is not encapsulated by the adhesive resin (17). The results of the present study revealed that changes in shear bond strength after thermal stress did not differ among the adhesive systems used. During thermal cycling, the specimens were subjected to stresses that were generated by differential thermal conductivity. The temperature changes that occur inside a specimen as a result of the thermal cycling are expected to have a significant impact (18). If collagen that is exposed by the acidic attack of a self-etch adhesive is insufficiently impregnated with resin components, hydrolysis might occur, leading to bonding failure (19). The thermal stress directed towards the bonding area might leave damaging residual defects or create new ones. Long-term water storage of bonded specimens has been reported to lead to degradation of the tooth-resin interface (20). Furthermore, the stress caused by thermal cycling might result in damage leading to surface crack initiation and propagation.

The cured layer of a single-step adhesive might act as a permeable membrane that allows water diffusion from the dentin to the interaction zone between the adhesive and the composite (21). Single-step self-etch adhesives have hydrophilic low-molecular-weight resin monomers that can infiltrate relatively deeply into the etched tooth substrate. Water movement across the cured adhesive layer might occur in the presence of low-molecular-weight resin monomers, allowing the diffusion of water from the hybridized dentin to the adhesive surfaces (22). Water diffusion into the bonding interface causes the resinous components to swell and become plasticized (23). During the thermal cycling test, hot water might accelerate the

Table 3 Effect of thermal cycling on shear bond strength (MPa) of single-step self-etch adhesive systems

Adhesive system	Storage conditions		
	24 h		10,000 TC
Adper Plompt L-Pop	10.4 (2.5)	*	11.3 (3.0)
(fracture mode)	9/1/0		10/0/0
AQ Bond Plus	9.3 (2.4)	*	9.6 (2.0)
(fracture mode)	9/0/1		10/0/0
Fluoro Bond Shake One	14.0 (2.9)	*	14.3 (3.0)
(fracture mode)	7/0/3		6/1/3
G-Bond	12.4 (1.5)	*	13.6 (1.9)
(fracture mode)	8/1/1		8/2 0
One-Up Bond F Plus	13.9 (2.6)	*	14.1 (1.5)
(fracture mode)	8/1/1		7/2/1

TC, thermal cycles; n = 10; values in parenthesis are SDs.
Fracture mode: adhesive failure/cohesive failure in resin/cohesive failure in dentin.
*: No significant differences between different storage conditions were found.
Values connected by lines are not significantly different (at a significance level $P > 0.05$)

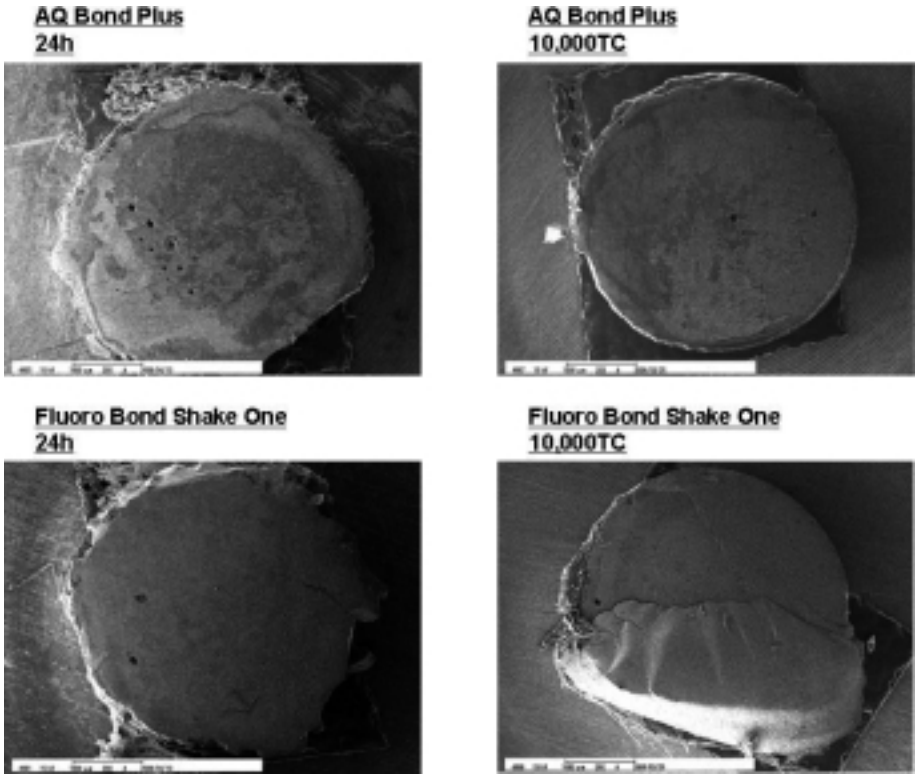

Fig. 1 Representative SEM observations of the fractured resin surface of a single-step self-etch system (AQ Bond Plus and Fluoro Bond Shake One) after 24 h and 10,000 thermal cycles. Bonding failure between the dentin and the adhesive is apparent (original magnification, ×20).

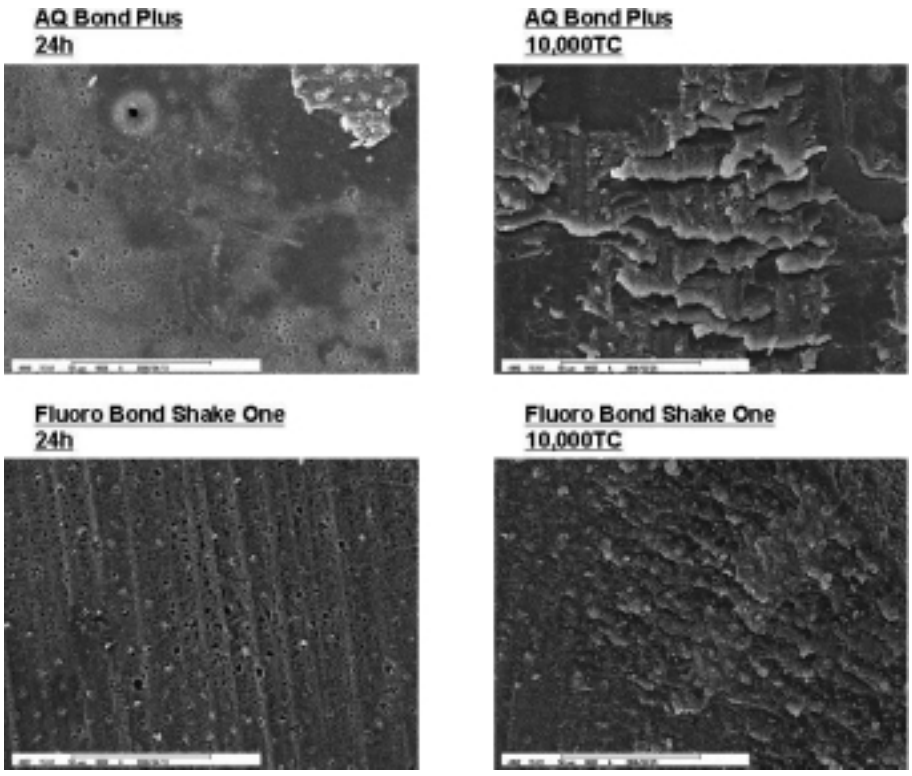

Fig. 2 Under higher magnification, several small cracks are visible on the cured adhesive resin (original magnification, ×800).

hydrolysis of the resin and extract poorly polymerized resin oligomers (24). The weakened mechanical properties of the composite resin might contribute to the decreased bond strengths of adhesive systems. Since single-step self-etch systems have a relatively thin interaction zone between dentin and adhesive, the detrimental effect of adverse environmental conditions might be less than that for adhesive systems utilizing strong acid with a thicker interaction zone.

A previous study that compared the chemical bonding efficacy of functional monomers demonstrated that a phosphate monomer had a high capacity for chemical bonding to hydroxyapatite over a clinically feasible application period. Furthermore, the calcium salt created by the phosphate monomer was highly insoluble. According to the adhesion-decalcification concept (25,26), the less soluble the calcium salt of an acidic molecule, the more intense and stable the molecular adhesion to a hydroxyapatite-based substrate. Thus the superior bonding performance of functional monomers might be reflected in their actual capacity for adhesive bonding to dentin after thermal cycling.

The results of this study suggest that use of single-step self-etch systems, in terms of simplifying clinical procedures, might have benefits even after exposure to a number of thermal cycles simulating conditions in the oral environment. However, it should be borne in mind that these results were obtained under *in vitro* conditions

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