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Influence of Thermal Cycling on Dentin Bond Strengths of Single-step Self-etch Adhesive Systems

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Clinical Relevance

Some of the bonding systems used in this study have achieved unchanged dentin bond strengths after thermal cycling. Therefore, by choosing appropriate adhesive systems, the simplified clinical procedures offered by single-step self-etch systems can be exploited even after thermal cycle stress.

SUMMARY

This study investigated the influence of thermal cycling on the dentin bond strengths of single-step self-etch adhesive systems. The Absolute, Clearfil Tri-S Bond, G-Bond and One-Up Bond F Plus systems were used. Bovine mandibular incisors were mounted in self-curing resin, and the facial surfaces were wet ground with #600 SiC paper. Adhesives were applied to the prepared dentin surfaces, and they were light irradiated according to each manufacturer's instructions. Resin composites were condensed into a mold (4 mm in diameter and 2 mm in height) and light irradiated for 30 seconds. Bonded specimens were divided into three groups and subjected to different storage conditions as follows: 37°C water for 24 hours; 37°C water for 24 hours followed by 10,000 thermal cycles between 5°C and 60°C or 37°C water for 24 hours followed by 20,000 thermal cycles between 5°C and 60°C. Ten samples per group were tested in a shear mode at a crosshead speed of 1.0 mm/minute. Analysis of variance and Tukey's HSD test at the 0.05 significance level were used to compare the three storage conditions for each adhesive system. After 24 hours storage in water, the mean dentin bond strengths ranged from 11.4 MPa to 17.1 MPa. The Clearfil Tri-S Bond system showed the highest bond strength. After 10,000 thermal cycles, the mean bond strengths remained unchanged

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except for those of the Clearfil Tri-S Bond system, which significantly increased. Significant decreases in bond strength were observed for the Absolute and One-Up Bond F Plus systems after 20,000 thermal cycles. Failure mode was commonly due to adhesive breakdown associated with partial cohesive failure of the adhesive resin. From the results of this study, in terms of simplifying the clinical procedure, the benefit of using single-step self-etch systems might be acceptable.

INTRODUCTION

New approaches to bonding restorative materials to tooth substrates without phosphoric-acid etching, such as self-etch systems, have recently been introduced.[1] These simplified systems aim to reduce the sensitivity of techniques by reducing the number of clinical steps involved. The use of single-step self-etch adhesives can prevent discrepancies occurring between the depth of etching and resin monomer penetration.[2] Single-step self-etch adhesive systems form a continuous layer by simultaneous demineralization with acidic monomers, followed by resin monomer penetration into the dentin substrate. There is no relationship between the depth of acid-etching by a self-etching primer and enamel strength.[3] By contrast, the application of self-etch adhesives to unprepared dentin resulted in a remaining smear layer and insufficient dentin bond strength.[4] This raises the question of whether the creation of a dentin/adhesive interaction zone might be sufficient to create stable adhesion after thermal cycle stress.[5-6]

The evaluation of bonding durability is important, as the bond between the restorative material and tooth substrate has a significant impact on the clinical success of tooth-colored restorations.[7] Although the most reliable conclusions about the performance of dental adhesive systems in the oral environment are derived from long-term clinical trials, the long-term storage of a bonded specimen in water, or subjecting the specimen 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.[9] Thermal cycling induces stress between a tooth substrate and a restorative material at the bonding interface. 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] Since the effect of thermal cycling on dentin bond strength is related to the bonding systems, an appropriate thermal challenge to the specimens should be considered. As the components and thickness of single-step self-etch adhesives differ from those of two-step adhesive systems, the thermal stresses created at the bonding interface might also vary.

This study determined the effect of thermal cycling on the dentin bond strengths of single-step self-etch adhesive systems to bovine dentin by measuring the shear bond strength and fracture mode and through scanning electron microscopy (SEM) observations of the fractured surface after the bond strength measurement.

METHODS AND MATERIALS

Adhesive Systems Tested

The following single-application self-etch adhesive systems, with a combination of resin composites, were used: Absolute/Esthet•X (Sankin Dentsply, Tokyo, Japan), Clearfil Tri-S Bond/Clearfil AP-X (Kuraray Medical, Tokyo, Japan), G-Bond/Gradia Direct (GC Corp, Tokyo, Japan) and One-Up Bond F Plus/Palfique Estelite Σ (Tokuyama Dental, Tokyo, Japan) (see Table 1 for details). 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, Danbury, 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.

Table 1: *Materials Tested*

Adhesive (manufacturer)	Main Components	Lot #	Restorative (shade)	Lot #
Absolute (Sankin Dentsply)	4-MET, PPTM, PEM-F, UDMA, initiator, and acetone	393-016	Esthet•X (Y-E)	0501132
Clearfil Tri-S Bond (Kuraray Medical)	MDP, bis-GMA, HEMA, initiator ethanol, stabilizer, and filler	040219	Clearfil AP-X (A2)	03841A
G-Bond (GC Corp)	4-MET, UDMA, acetone, water silanated colloidal silica, and initiator	0403191	Gradia Direct (A2)	0312121
One-Up Bond F Plus (Tokuyama Dental)	MAC-10, HEMA, MMA, multifunctional methacrylic monomer fluoroaluminosilicate glass, and water photo-initiator (aryl borate catalyst)	A: 003 B: 504	Palfiqu Estelite Σ (A2)	M2J01184S

4-MET, 4-methacryloyloxyethyl trimellitate; bis-GMA, 2,2bis[4-(2-hydroxy-3-methacryloyloxypropoxy)phenyl] propane; HEMA, 2-hydroxyethyl methacrylate; MAC-10, 11-methacryloyloxy-1, 1-undecanedicarboxylic acid; MDP, 10- methacryloyloxydecyl dihydrogen phosphate; PEM-F, fluoromethacryloxy cyclophosphazen; PPTM, pyrophosphate tetramethacrylate; UDMA, urethane dimethacrylate.

Bond-strength Test

Mandibular incisors extracted from two- to three-year-old cattle and stored frozen for up to 2 weeks were used as a substitute for human teeth.[13-15] 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 they were 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 one minute to remove any excess debris, the surfaces were washed and dried with oil-free compressed air.

Table 2: *Application Protocols for Single-step Self-etch Adhesive Systems*

Adhesive System	Application Protocol
Absolute (1 bottle)	Dispense 1 drop of liquid into a well. Apply to dentin for 5 seconds with moderate finger pressure. Apply a gentle stream of air to dry, and then apply a second coat of adhesive. Gently air dry for 3 seconds and then light irradiate for 10 seconds.
Clearfil Tri-S Bond (1 bottle)	Dispense 1 drop of liquid into a well. Apply to dentin for 20 seconds. Apply a relatively strong stream of air to dry and then light irradiate for 10 seconds.
G-Bond (1 bottle)	Apply sufficient adhesive for 10 seconds. Use a strong stream of air to dry and then light irradiate for 10 seconds.
One-Up Bond F Plus (2 bottles)	Mix equal amounts of the bond agents A and B until a pink homogenous liquid mixture is obtained. Apply to dentin for 10 seconds with agitation and then light irradiate for 10 seconds.

Table 3: *Influence of Thermal Cycling on Dentin Bond Strength (MPa) in Single-step Self-etch Adhesive Systems*

	Storage Conditions		
Adhesive System	**24 Hours**	**10,000 TC**	**20,000 TC**
Absolute	12.8 (1.9)[a]	13.8 (1.7)[a]	8.2 (1.4)[b]
(fracture mode)	10/0/0	8/2/0	8/2/0
Clearfil Tri-S Bond	17.1 (1.7)[c]	22.6 (2.1)[d]	20.4 (2.2)[d]
(fracture mode)	7/2/1	6/3/1	7/2/1
G-Bond	11.4 (1.5)[e]	10.9 (1.9)e	11.8 (3.0)[e]
(fracture mode)	8/1/1	8/2 0	9/1/0
One-Up Bond F Plus	14.3 (2.6)[f]	14.1 (1.5)[f]	9.0 (1.9)[g]
(fracture mode)	8/1/1	7/2/1	8/1/1

TC, thermal cycles; n=10; values in parenthesis are SDs. Fracture mode: adhesive failure/cohesive failure in resin/cohesive failure in dentin. Values within the same rows denoted by superscript letters are not significantly different at a significance level $p>0.05$.

To define the designated bonding area, a piece of double-sided adhesive tape, bearing a hole 4 mm in diameter, was firmly attached. The adhesive was applied to the dentin surface according to each manufacturer's instructions (Table 2). After the adhesive had been applied, the surfaces were dried with oil-free compressed air that was administered at a pressure of 0.2 MPa from 5 cm above the dentin surface for five seconds 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, then hold, the restorative resin on the dentin surface. The resin composite was condensed into the mold and cured for 30 seconds. The Teflon mold and adhesive tape were removed from the specimens 10 minutes after light irradiation. The bonded specimens from each set of materials were divided into three treatment groups (n=10 in each) as follows: group 1 specimens were stored in 37°C water for 24 hours after placement without thermal cycling; group 2 specimens were stored in 37°C water for 24 hours, followed by 10,000 thermal cycles between 5°C and 60°C and group 3 specimens were stored in 37°C water for 24 hours, followed by 20,000 thermal cycles between 5°C and 60°C. The dwell time in the water bath was 30 seconds and the transfer time was five seconds.

After treatment, the specimens in each group were tested in shear mode using a knife-edge testing apparatus in a universal testing machine (Type 4204, Instron Corp, Canton, MA, USA) at a crosshead speed of 1.0 mm/minute. 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 10x magnification in order to identify the location of the bond failure. The type of bond failure was determined based on the percentage of substrate-free material as follows: adhesive failure, cohesive failure of the resin (adhesive or composite) or cohesive failure of the dentin.[15]

Statistical Analyses

The results were analyzed by calculating the mean shear bond strength (MPa) and standard deviation (SD) for each group. Statistical tests were performed to determine how thermal cycling influenced bond

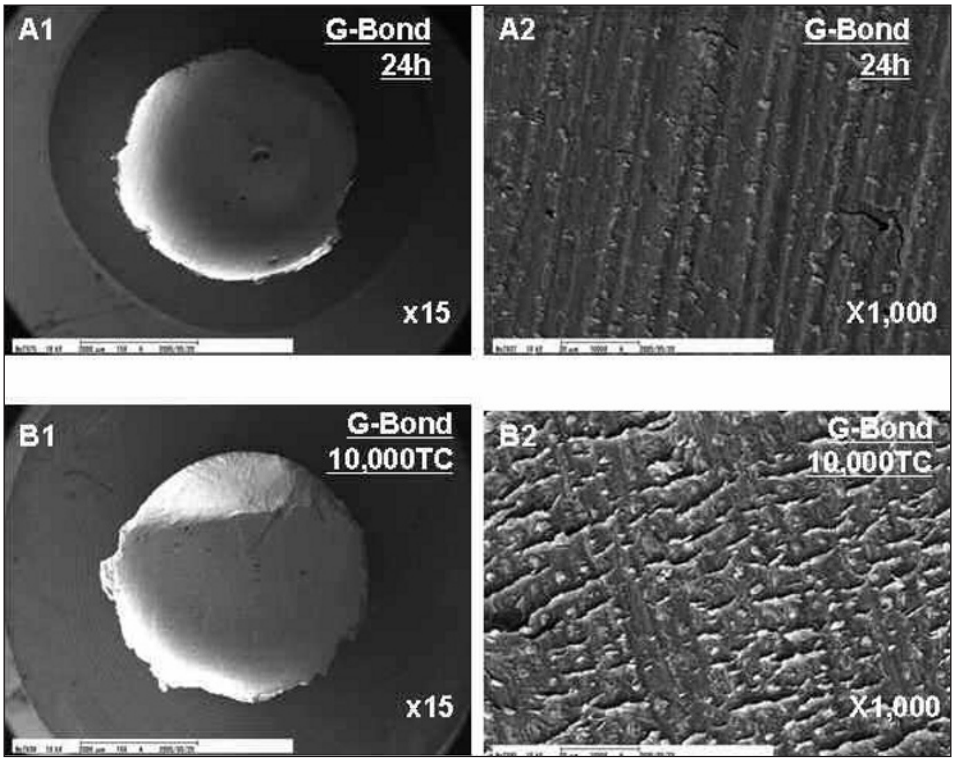

Figure 1: *Representative SEM observations of the fractured resin surface of a single-step self-etch system (G-Bond) after 24 hours and 10,000 thermal cycles. Bonding failure between the dentin and adhesive is apparent (A1, B1). Under higher magnification, several small cracks are visible on the cured adhesive resin (A2, B2).*

strengths. The data for each group within each adhesive system were tested for homogeneity of variance using Bartelett's test, then subjected to analysis of variance (ANOVA) followed by Tukey's HSD test at the 0.05 significance level. All statistical analyses were carried out using the Sigma Stat Ver 3.1 (SPSS Inc, Chicago, IL, USA) software system.

SEM

The ultrastructure of the dentin surfaces was examined by field-emission (FE)-SEM (ERA-8800FE, Elionix Ltd, Tokyo, Japan). Briefly, fractured specimens were dehydrated after the bond strength measurements in ascending concentrations of *tert*-butanol (50% for 20 minutes, 75% for 20 minutes, 95% for 20 minutes and 100% for 2 hours), then transferred to a critical-point dryer for 30 minutes. The surfaces were coated in a vacuum evaporator, Quick Coater (Type SC-701, Type SC-701, Sanyu Denshi Inc, Tokyo, Japan) with a thin film of gold. The specimens were then observed under FE-SEM.

RESULTS

Table 3 shows the mean shear bond strengths to bovine dentin and the causes of failure after the testing (statistical significance is denoted using superscript letters). After 24 hours storage in water, the mean dentin bond strengths of the single-step self-etch systems ranged from 11.3 MPa to 16.9 MPa. The Clearfil Tri-S Bond system numerically showed the highest bond strengths at each storage condition. Significant decreases in bond strength after 20,000 thermal cycles were observed for the Absolute and One-Up Bond F Plus systems, while significant increases in bond strength were observed for the Clearfil Tri-S Bond after thermal stress. For G-Bond, no changes in bond strength were found in each storage condition.

After 24 hours storage in water, the predominant modes of failure are shown as adhesive breakdown at the dentin surface associated with partial cohesive failure of the resin. The FE-SEM observations of the fractured resin surfaces (Figure 1) revealed adhesive failure between the dentin and adhesive (Figure 1: A1, B1). In addition, several small cracks were observed on the cured adhesive resin under higher magnification (Figure 1: A2, B2).

DISCUSSION

The authors' previous study[16] of two-step self-etch adhesive systems suggested that dentin bond strength decreased as the number of thermal cycles increased; this could be attributed to the presence of a region of demineralized dentin that was not encapsulated by the adhesive resin.[17] Theoretically, adhesive penetrates the etched dentin simultaneously and hardens after evaporation of the solvent and exposure to light.[2] This process creates a thin interaction zone and mechanical retention between the dentin and resin composite.

The use of thermal cycling is a method of providing laboratory stimulations of oral conditions. The number of thermal cycles and the temperatures used are different among bond strength studies. From a literature review[9] on the durability of adhesives to tooth structure, 10,000 thermal cycles corresponds to approximately one year of *in vivo* functioning. The fact that the 20,000 thermal cycles used in this study significantly decreased shear bond strengths for some adhesive systems might indicate that 20,000 cycles was an appropriate thermal cycle for the specimens.

The results of this study revealed that changes in shear bond strength after thermal cycling differ among single-step self-etch adhesive systems. During thermal cycling, the specimens were subjected to mechanical stresses that were generated by differential thermal conductivities. It has been speculated that, in large specimens subjected to thermal cycling, a significant portion of the interface might be thermally protected.[8] However, temperature changes that occur inside a specimen as a result of the temperature of the surrounding water are expected to have a significant impact.[18] In this study, a sequence of temperature changes from 5°C to 55°C, with a corresponding dwell time of 30 seconds, was chosen, as this was believed to represent a suitably discriminatory challenge.[9]

Specimen geometry is one of the key factors when considering the effect of storage conditions on bond strength. Indeed, one explanation for a lack of change in bond strength after thermal stress is a low C-factor, which might explain why differential behavior does not alter the interfacial bond.[19] Here, the resin column was bonded to the flat tooth surface in the specimens used to test shear bond strength. It is possible, therefore, that a slight contraction stress was generated at the bonding interface due to the differential coefficient of thermal expansion. As long-term water storage of bonded specimens has been reported to lead to degradation of the tooth–resin interface,[20] the stress caused by thermal cycling might result in damage leading to crack initiation and propagation at this surface. Thus, in order to subject the bonded specimens to relatively aggressive thermal stress, up to 20,000 thermal cycles were employed in the current study. Although it is impossible to stimulate chemical attack from water into the bonding interface, thermal cycling is one of the methods to assess the effect of prolonged water exposure on dentin bond strengths.

The cured layer of a single-step adhesive might act as a permeable membrane that allows water diffusion from the hydrated dentin to the intermixed zone between the adhesive and composite.[21] Single-step self-etch adhesives have low molecular weight resin monomers that are hydrophilic in nature and 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 created by an adhesive and a tooth substrate caused the resinous components to swell and become plasticized.[23] During the thermal cycling test, hot water might accelerate hydrolysis of the resin and extract poorly polymerized resin oligomers.[24] The weakened mechanical properties of the resin composite might contribute to the decreased bond strengths of the adhesive systems. In fact, failure after thermal cycling tended to result from the weakened resin between dentin and the restorative material.

If collagen that is exposed by the acidic attack of self-etch adhesive is insufficiently impregnated with resin components, hydrolysis might occur, leading to bonding failure.[25] Thermal stress directed towards the bonding area might create defects or damaging residual defects. SEM observations of the fractured specimens after the test suggested that the durability of the bonded joint was threatened by thermal stress. The most susceptible region was the dentin–resin interface. According to the *in vivo* study that investigated the degradation of resin-dentin bond structures, the complete loss of resinous materials between collagen fibrils on depletion of collagen fibrils within the degraded hybridized dentin were observed.[26] Deterioration of the resin-dentin bond structure might be caused by the creation of nano-sized water diffusion, resulting in lower bond strengths for some systems used in this study.

A previous study that compared the chemical bonding efficacy of the functional monomers 10-methacryloyloxydecyl dihydrogen phosphate (MDP), 4-methacryloxyethyl trimellitic acid (4-MET) and 2-methacryl oxyethyl phenyl hydrogen phosphate (phenyl-P) reported that MDP had a high chemical bonding potential to hydroxyapatite over a clinically feasible application period.[27] Furthermore, the calcium salt of MDP was highly insoluble and, consequently, could not withstand ultrasonic cleaning. According to the adhesion–decalcification concept,[28] the less soluble the calcium salt of an acidic molecule, the more intense and stable the molecular adhesion to a hydroxyapatite-based substrate. The superior bonding performance of MDP might have been reflected in the actual adhesive potential to dentin after thermal cycling. Accordingly, an increase in bond strength was observed for the Clearfil Tri-S Bond system after thermal cycling.

CONCLUSIONS

The results of this study suggest that the benefits of using single step self-etch systems, in terms of simplifying clinical procedures, might be viable even after exposure to a number of thermal cycles simulating the oral environment. Care should be taken, however, as these results were obtained under laboratory conditions, and it remains to be seen whether a similar trend will occur *in vivo*. It is crucial for general practitioners who use these adhesive systems to understand the factors that contribute to the durability of the restorations and to be aware of their bonding characteristics.

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