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Effect of Thermal Cycling on Enamel Bond Strength of Single-step Self-etch Systems

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Clinical Relevance

After thermal cycling, some recently introduced simplified bonding systems showed no change in enamel bond strengths.

SUMMARY

This study investigated the influence of thermal cycling on the enamel bond strength of single-step self-etch adhesive systems.

The systems used were Absolute, Clearfil tri-S Bond, G-Bond and One-Up Bond F Plus. Bovine mandibular incisors were mounted in self-curing resin, and the facial surfaces were wet ground with #600 SiC paper. Adhesives were applied on the prepared enamel surfaces and light irradiated according to each manufacturer's instructions. Resin composites were condensed into a mold (ø4x2 mm) and light irradiated for 30 seconds. Thirty specimens per adhesive systems were divided into 1 of 3 test groups (n=10) following storage in water at 37°C for 24 hours. The specimens were then stored in 37°C water for 24 hours, followed by thermal cycling 10,000 and 20,000 times between 5°C and 60°C. After each storage condition, the specimens were tested in shear mode at a crosshead speed of 1.0-mm/minute. One-way ANOVAs and Tukey HSD test at a level of 0.05 were conducted.

After 24 hours of water storage, the mean enamel bond strengths ranged from 11.3 to 16.9 MPa, and Clearfil tri-S Bond showed significantly higher bond strength. After thermal cycling, the mean bond strengths ranged from 8.3 to 20.7 MPa. The changes in enamel bond strengths were different among the adhesive systems tested. Failure modes after the test were commonly adhesive failure associated with partial cohesive failure adhesive in resin.

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With a careful choice of adhesive systems, the benefit to using single-step self-etch systems in terms of simplifying the clinical procedure might be acceptable, even after thermal stresses.

INTRODUCTION

Using the acid-etch technique to modify enamel structure with phosphoric acid[1] has become a standard procedure for the surface conditioning of enamel prior to adhesive resin application. The retentive ability of composite to etched enamel is described as a function of an increase in bonding area and wettability of the adherend surface.[2-4] Infiltration of the adhesive resin into the porous zone resulted in the formation of resin tags, thus establishing micromechanical retention to etched enamel. New approaches of bonding to tooth substrate without phosphoric acid etching, such as self-etch systems, have been introduced.[5] These simplified systems are suggested to reduce technique sensitivity and shorten clinical procedures. The use of single-step self-etch adhesives may eliminate possible discrepancies between depth of etching and resin monomer penetration.[6]

Single-step self-etch adhesive systems form a continuous layer by simultaneous demineralization with acidic monomers followed by resin monomer penetration into the enamel surface. Penetration of these acidic monomers into etched enamel creates resin tags. Although no relationship between depth of acid-etching of self-etching primer and bond strength has been shown,[7] the application of self-etch adhesives to unprepared enamel resulted in a shallow etching pattern and revealed insufficient bond strengths.[8-9] On the other hand, it has been reported that a comparatively higher bond strength was obtained for self-etching primer adhesive systems with the creation of a submicron resin tag.[10] The question arises whether the resulting bonds might be stable after thermal cycle stressing.[11]

With tooth colored restorations, evaluation for bonding durability is important, since stability of the bond between the restoration and tooth substrate is important for long-term clinical success.[12] Although the most reliable conclusions regarding the performance of dental adhesive systems in the oral environment must be derived from long-term clinical trials, long-term storage of the bonded specimen in water, or subjecting dental adhesive systems to thermal cycling, may provide some information about temperature dependent degradation of the material.[13] A thermal cycling test is the process of subjecting specimens to extreme temperatures that simulate intraoral conditions.[14] Also, this test induces stress between tooth substrate and restorative materials due to differences in their coefficients of thermal expansion. It is reported that the effect of thermal cycling on the bond strength of multi-step bonding systems depended on the bonding systems used and the number of thermal cycles.[13,15-17] Since single-step self-etch adhesive interfaces are thinner than those created by 2-step self-etch systems, thermal stresses created at the bonding site might be different.

This study determined the effect of thermal cycling on the enamel bond strengths of single-step self-etch adhesive systems to bovine enamel by means of measurement of shear bond strength, fracture mode and field emission scanning electron microscopy (FE-SEM) observations of the fractures' surface after bond strength measurements. The null hypothesis to be tested was that thermal cycling would not affect bond strength to bovine enamel.

METHODS AND MATERIALS

Materials Tested

Single-step self-etch adhesive systems with the following combinations of resin composites that were used include: 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/Estelite ∑ (Tokuyama Dental, Tokyo, Japan) (Table 1). All the adhesive systems were used in combination with the manufacturers' restorative resins. 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, and the power density (800 mW/cm^2) of the light was checked with a dental radiometer (Model 100, sds Kerr) before making specimens.

Bond Strength Test

Mandibular incisors extracted from 2-3 year old cattle and stored frozen (-20°C) for up to 2 weeks were used as a substitute for human teeth. After removing the roots of the teeth with a slow-speed saw and 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 a flat enamel surface. Each tooth was then mounted in self-curing acrylic resin (Trey Resin II, Shofu Inc, Kyoto, Japan) to expose the flattened area and placed in tap water to reduce the temperature rise from the exothermic polymerization reaction of the acrylic resin. The final finish was accomplished by grinding on wet 600-grit SiC paper. After ultrasonic cleaning with distilled water for 1 minute to remove excess debris, these surfaces were washed and dried with oil-free compressed air.

A piece of double-sided adhesive tape with a hole 4-mm in diameter was firmly attached to the flat enamel

Table 1: *Materials Tested*

Adhesive (Manufacturer)	Main Components	Lot #	Restorative (Shade)	Lot #
Absolute (Sankin Dentsply)	4-MET, PPTM, PEM-F, UDMA, initiator, acetone	393-016	Estet•X (Y-E)	0501132
Clearfil tri-S Bond (Kuraray Medical)	MDP, bis-GMA, HEMA, initiator, ethanol, stabilizer, filler	040219	Clearfil AP-X (A2)	03841A
G-Bond (GC Corp)	4-MET, UDMA, acetone, water, silanated colloidal silica, initiator	0403191	Gradia Direct (A2)	0312121
One-Up Bond F Plus (Tokuyama Dental)	MAC-10, HEMA, MMA, multifunctional methacrylic monomer, fluroaluminosilicate glass, water, photoinitiator (aryl borate catalyst)	A: 003 B: 504	Estelite∑ (A2)	2J01184S

4-MET: 4-methacryloyloxyethyl trimellitate , PPTM: pyrophosphate tetramethacrylate, PEM-F: fluoromethacryloxy cyclophosphazen, UDMA: urethane dimethacrylate, MDP: 10-methacryloxydecyl di-hydrogen phosphate, bis-GMA: 2, 2bis[4-(2-hydroxy-3-methacryloyloxypropoxy)phenyl] propane, HEMA: 2-hydroxyethyl methacrylate, MAC-10: 11-methacryloxy-1,1-undecandicarboxylic acid

Table 2: *Application Protocols of Single-Application Adhesive Systems*

Adhesive System	Application Protocol
Absolute (Single Bottle)	Dispense one drop of liquid into well. Apply to enamel for 5 seconds with moderate finger pressure. Gentle stream of air to dry and apply second coat of adhesive. Gently air dry for 3 seconds and light irradiation for 10 seconds.
Clearfil tri-S Bond (Single Bottle)	Dispense one drop of liquid into well. Apply to enamel for 20 seconds. Relatively strong stream of air to dry and light irradiation for 10 seconds.
G-Bond (Single Bottle)	Apply sufficient amount of adhesive for 10 seconds. Strong air dry and light irradiation 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 enamel for 10 seconds with agitation and light irradiation for 10 seconds.

surface to define the area for bonding. The adhesive was applied on the enamel surface according to the manufacturer's instructions (Table 2). The surfaces with the adhesive were dried for 5 seconds with oil-free compressed air at 0.2 MPa pressure from 5 cm above the enamel surface using a 3-way syringe; they were then irradiated with the curing unit. A Teflon (Sanplatec Corp, Osaka, Japan) mold, 2.0-mm high and 4.0-mm in diameter, was used to form and hold the restorative resin onto the enamel 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.

Bonded specimens from each group of materials were divided into 3 treatment groups of 10 specimens each for testing: Group 1) stored in 37°C distilled water for 24 hours after placement without thermal cycling, Group 2) stored in 37°C distilled water for 24 hours, followed by thermal cycling 10,000 times between 5°C and 60°C and Group 3) stored in 37°C distilled water for 24 hours, followed by thermal cycling 20,000 times. The dwell time in each bath was 30 seconds and the transfer time was 5 seconds.

The specimens in each group were tested in shear mode using 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 in MPa were calculated from the peak load at failure divided by the specimen surface area.

After testing, the specimens were examined in an optical microscope (SZH-131, Olympus Ltd, Tokyo, Japan) at 10x magnification to define the location of the bond failure. The type of failure was determined based on the percentage of substrate-free material: adhesive failure, cohesive failure in composite/adhesive resin with partial adhesive failure (mixed mode) and cohesive failure in enamel.[18]

Statistical Analysis

The results were analyzed by calculating the mean shear bond strength (MPa) and standard deviation for each group. A statistical analysis was done to show how the bond strengths were influenced by thermal cycling. The data for each group were tested for homogeneity of variance using Bartelett's test, then subjected to 1-way ANOVA followed by the Tukey HSD test at α=0.05 within each adhesive system. The statistical analysis was carried out with the Sigma Stat software system (Ver 2.01, SPSS Inc, Chicago, IL, USA).

Table 3: *Influence of Thermal Cycling on Bond Strength (MPa) of Single-step Bonding Systems to Bovine Enamel*

Adhesive System	24 hours	10,000 TC	20,000 TC
Absolute	11.3 (3.2)[a]	11.1 (1.9)[a]	8.3 (3.5)[b]
Clearfil tri-S Bond	16.9 (1.8)[c]	20.1 (2.4)[d]	20.7 (1.6)[d]
G-Bond	12.0 (2.0)[e]	15.0 (2.6)[f]	13.2 (3.0)[e,f]
One-Up Bond F Plus	12.6 (1.8)[g]	11.8 (1.8)[g]	13.4 (3.4)[g]

N=10
TC: Thermal cycling
Groups within the same row with the same superscript letters are not significantly different (p>0.05)

Scanning Electron Microscopy

For ultrastructure observation of the enamel surfaces by FE-SEM, after bond strength measurements, the fractured specimens were dehydrated 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, Sanyu Denshi Inc, Tokyo, Japan) with a thin film of gold. The specimens were then observed using FE-SEM (ERA-8800FE, Elionix Ltd, Tokyo, Japan).

RESULTS

The mean shear bond strengths to bovine enamel are shown in Table 3. Alphabetic characters are used in the tables to show the results of the statistical analysis. After 24 hours water storage, the mean enamel bond strengths of the single-step self-etch systems ranged from 11.3 to 16.9 MPa. Changes in mean enamel bond strength were different among the adhesive systems. A significant decrease in bond strength after 20,000 thermal cycles was observed for Absolute, an increase in bond strength was noted for Clearfil tri-S Bond and no changes in bond strength were seen for One-Up Bond F Plus.

Figure 1 shows the result of the representative FE-SEM observations of fractured resin surfaces of 10,000 thermal cycle subjected specimens. The failure mode after the bond strength test was not uniform. Mixed mode, including adhesive failure at the resin-enamel interface and partial cohesive failure in adhesive resin, were most commonly recorded for all specimens except Absolute. Absolute showed adhesive failures at the resin-enamel interfaces, regardless of subjection to thermal cycling. Cohesive failure in enamel was not recorded in any of the adhesive systems. More

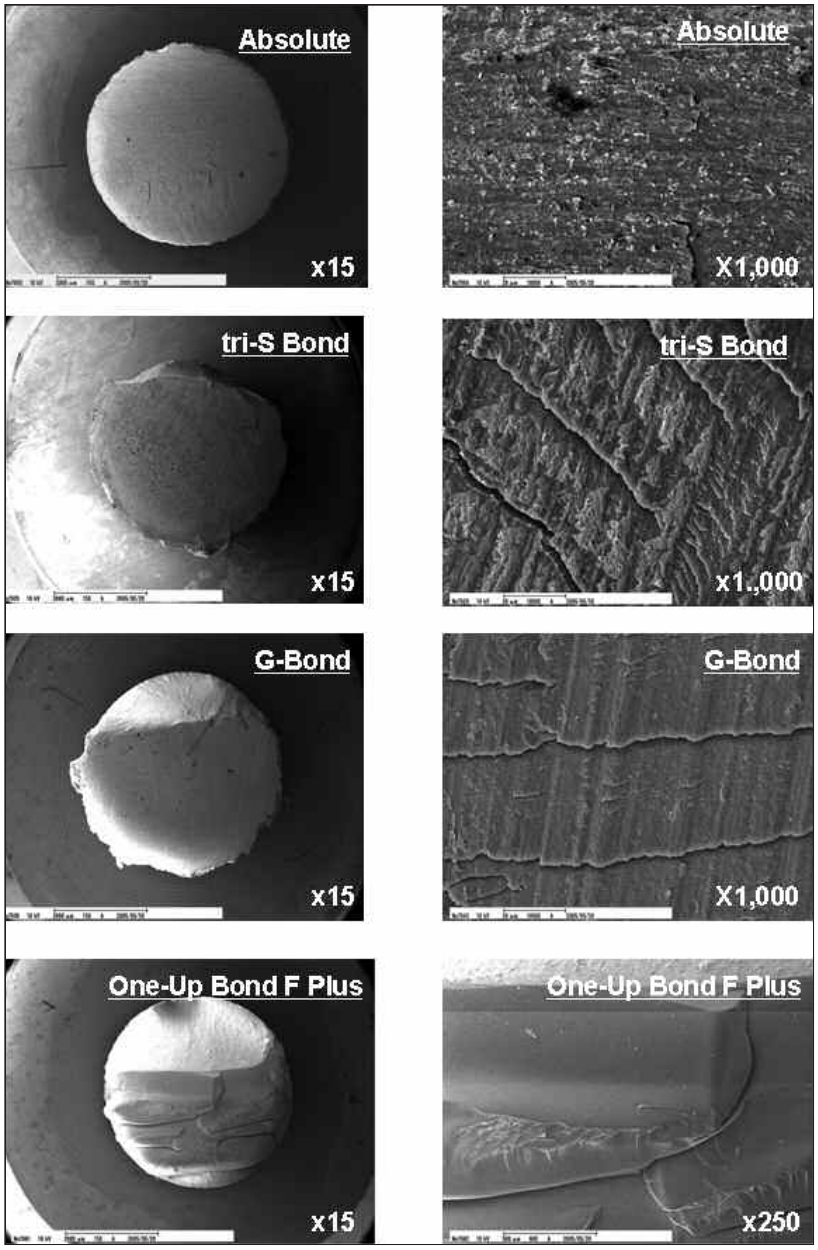

Figure 1*: FE-SEM observations of fractured resin surface of single-step self-etch systems after 10,000 times thermal cycling. Adhesive failure between enamel and adhesive is observed for Absolute. With higher magnification, several small cracks or cleavages are observed on the cured adhesive resin for the adhesive systems except for Absolute.*

cleavage steps, typical for the fracture pattern of brittle materials, were observed on fractured adhesive resin surfaces after subjecting them to thermal cycling than for the 24-hour storage specimens.

DISCUSSION

The depth of surface enamel removed during the etching procedure depends on the type of acid, concentration, duration of etching and chemical composition of the surface enamel.[19] The authors' previous study on the durability of enamel bond strength of 2-step self-etch adhesive systems suggest that bond strengths tend to decrease with the number of thermal cycles,[20] which might be evidence of a region of demineralized enamel that is not penetrated by adhesive resin.[21] Theoretically, the adhesive penetrates the etched enamel and hardens after evaporation of the solvent and light exposure. This process creates an enamel-resin interaction zone with stable resin tags and mechanical retention between the enamel and resin composite. The question is whether single-step self-etch adhesives are capable of providing sufficient bonding to enamel such that a stable micromechanical bond can be achieved.

The hypothesis that thermal cycling would not affect enamel bond strengths was rejected for some of the adhesive systems tested. Based on the results of this study, changes in enamel bond strength after thermal cycling varied among single-step self-etch adhesive systems, and relatively small changes in bond strengths were recorded, except for Absolute. During thermal cycling, the specimens were subjected to mechanical stresses generated by differential thermal expansion and conductivity. It has been speculated that specimen size subjected to thermal cycling might be so great that the specimen mass is thermally protected, because it might be limited by surrounding tooth and composite.[13] However, it has been reported that the temperature change inside the specimen, based on changes in the surrounding water temperature, would have more than a superficial effect.[22] In this study, the sequence of temperatures, 5°C and 55°C, with a corresponding dwell time of 30 seconds, has been chosen, as recommended, to be a suitable discriminatory challenge.[14]

Specimen geometry is one of the key factors when considering the effect of storage conditions on bond strength. It also serves as another explanation for small changes in bond strength after thermal stress is the low C-factor, which may explain why differential expansion would not alter the interfacial bond.[23] Since the resin column was bonded to a flat tooth surface for shear bond strength specimens, low contraction stresses might be generated at the bonding interface, due to the differential coefficient of thermal expansion. Long-term water storage of the bonded specimen was also reported to lead to degradation of the tooth-resin interface,[24] and flaws caused by thermal stress might result in damage that leads to crack initiation and propagation at the bonding interface. To create more thermal stress cycles on bonded specimens, up to 20,000 thermal were employed in this study. Although degradation in enamel substrate could occur, its effect on bond strength was not demonstrated for most of the adhesive systems studied.

Single-step self-etch adhesives have low molecule weight resin monomers that exhibit a relatively hydrophilic nature.[25-26] Incorporating high concentrations of acidic monomers might lead to water sorption, resulting in a decline in the marginal integrity of the adhesives. Water diffusion into the bonding interface created by adhesive and tooth substrate causes resin components to swell and plasticize.[27-28] The complex thermal cycling process offers many possibilities for the entrapment of flaws inside the enamel-resin interface.[20] The thermal cycling test induces stress between the tooth substrate and restorative materials due to differences in their coefficients of thermal expansion. During the thermal cycling test, hot water may accelerate the hydrolysis of resin and extract poorly polymerized resin oligomers.[29-30] Water and other chemicals leaching from the oral cavity might decrease the mechanical properties of polymers. The decreased mechanical properties of resin composite might contribute to the decreased bond strengths of any adhesive systems. The change in mechanical properties after thermal cycling could result in a tendency toward bond failure due to weakened resin tags, which exist between etched enamel and resin.

Specimens showing higher bond strength tended to have rougher fracture surfaces. In specimens subjected to thermal cycling, a river pattern, or cleavage steps, appeared on the fractured surface for the adhesive systems that showed higher bond strengths.[31] The increased fracture resistance of adhesive resin is possibly through crack pinning, crack branching by toughening and plastic deformation of the resin component. Therefore, more energy is needed for crack propagation.[32] Microcracks observed inside adhesive resins might also increase fracture resistance of the materials—the so-called microcrack-induced toughening effect.[33]

From a previous study that compared the chemical bonding efficacy of functional monomers (MDP; 10-methacryloxydecyl dihydrogen phosphate, 4-MET; 4-methacryloxyethyl trimellitic acid and phenyl-P; 2-methacryloxyethyl phenyl hydrogen phosphate), MDP has been reported to have a high chemical bonding potential to hydroxyapatite within a clinically reasonable application time.[34] Furthermore, the calcium salt of MDP was highly insoluble and, consequently, was able to resist ultrasonic cleaning. According to the adhesion-decalcification concept,[35] the less soluble the calcium salt of the acidic molecule, the more intense and stable the molecular adhesion to a hydroxyapatite-based substrate. The adhesive potential of MDP might be reflected in the higher, more stable bonding performance to enamel after thermal cycling. This could explain the increase in bond strength observed for Clearfil tri-S Bond after thermal cycling.

The results of this study suggest that the benefit of using single-step self-etch systems in terms of simplifying the clinical procedure might be acceptable even after a number of thermal cycles attempt to simulate long-term exposure to the oral environment. The general practitioners who use these adhesive systems should be aware, however, that 1 of the 4 single-step self-etch systems studied demonstrated a significant decrease in bond strength after thermal cycling. A further understanding of the factors that contribute to the durability of the restorations and their bonding characteristics is needed, and, for comparison, it would be helpful to conduct bond strength testing using traditional multi-step adhesive systems as a control.

CONCLUSIONS

The influence of thermal cycling on the enamel bond strengths of single-step self-etch systems was examined, and the following conclusions can be drawn.

1. All but 1 of the 4 adhesive systems used in this study showed no significant decrease in bond strength after up to 20,000 cycles of thermal stress. One system actually demonstrated a significant increase in bond strength after cycling.
2. The adhesives whose bond strengths were not decreased by thermal cycling showed mixed adhesive-cohesive failure at the interface.
3. From scanning electron microscopy observations on fractured resin surfaces, more cleavage steps, which are typical of the fracture pattern of brittle materials, were observed on the fractured adhesive resin surfaces in specimens subjected to thermal cycling.
4. Further study is needed to better understand the behavior of self-etching adhesive monomers after thermal stressing.

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