

## Enamel Bonding of Self-etch and Phosphoric Acid-etch Orthodontic Adhesive Systems

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This study was conducted to examine the shear bond strengths of orthodontic brackets to ground enamel with two self-etching adhesives (Beauty Ortho Bond and Transbond XT) and two phosphoric acid-etching adhesives (Kurasper F and Orthomite Superbond). After the bonded specimens were stored in water at 37 °C for 24 hours, they were subjected to thermal cycling (TC) at 0 and 5000 cycles. Then, shear bond test was performed at a crosshead speed of 1 mm/min. Results indicated that the self-etching adhesives did not significantly differ in bond strength before and after TC ( $p > 0.05$ ), while the phosphoric acid-etching adhesives showed a significant reduction after TC ( $p < 0.05$ ). The predominant modes of bracket failure for the self-etching adhesives were at the enamel-resin interface, leaving less than 50% of the adhesive on the enamel surface. It was concluded that the self-etching adhesives exhibited stable bond strength to ground enamel without causing decalcification nor damage to the enamel surface.

Keywords: Orthodontic brackets, Shear bond strengths, Self-etching primer

### INTRODUCTION

Phosphoric acid etching of enamel was introduced by Buonocore<sup>1</sup> in 1955, which has since led to dramatic changes in the practice of orthodontics<sup>2</sup>. By the 1970s, the bonding of orthodontic brackets had become an accepted clinical technique<sup>3,4</sup>. Bonded orthodontic brackets have advantages over bands in that they have no interproximal contacts, are easier to place and remove, are more esthetic, hygienic, and less irritating to the gingiva<sup>5</sup>.

However, the components of the appliance and the bonding materials often promote plaque accumulation with subsequent acid production, leading to decalcification and an alteration in the appearance of the enamel surface<sup>6</sup>.

Although the acid etching technique is a useful procedure in orthodontics, there is a need to improve the bonding procedure for two key reasons: to maintain clinically useful bond strengths while minimizing the amount of enamel loss, and to simplify the technique by reducing the number of steps.

Bonding systems used in operative dentistry were developed to combine conditioning and priming agents into a single acidic primer for simultaneous use on enamel and dentin, eliminating the separate steps of etching, rinsing, and drying<sup>7,8</sup>. The use of a self-etching primer offers the advantage of a faster and simplified application technique, thereby allowing adequate etching and priming of enamel and dentin in only one step<sup>9</sup>. In addition to saving time, fewer steps in the bonding process might translate into

fewer procedural errors, thus minimizing technique sensitivity.

A self-etching primer system has been introduced for the bonding of orthodontic brackets<sup>10</sup>. Bishara *et al.*<sup>11</sup> reported that the use of a self-etching primer system resulted in a clinically acceptable bond strength.

Recently, a new orthodontic adhesive, Beauty Ortho Bond (Shofu, Kyoto, Japan), was developed, which is composed of a self-etching primer and a fluoride-releasing adhesive system.

The purpose of this study was to examine the shear bond strengths of self-etch and phosphoric acid-etch orthodontic adhesive systems to enamel. In addition, the modes of bracket failure were examined using scanning electron microscopy (SEM).

### MATERIALS AND METHODS

#### *Materials used in this study*

The materials used in this study are listed in Table 1. Two self-etch adhesive systems, Beauty Ortho Bond (BO, Shofu, Kyoto, Japan) and Transbond XT (TB, 3M Unitek, Monrovia, CA, USA), and two phosphoric acid-etch adhesive systems, Kurasper F (KF, Kuraray Medical, Tokyo, Japan) and Orthomite Superbond (OS, Sun Medical, Moriyama, Japan), were used in this study.

Beauty Ortho Bond is composed of a self-etching primer and a fluoride-releasing light-cured adhesive system. The primer includes a phosphonic acid monomer, which contributes to etching enamel. The

Table 1 Materials used in this study.

Material	Manufacturer	Batch No.	Composition	Instructions
Beauty Ortho Bond (BO)	Shofu, Kyoto, Japan	Primer A: 11031301	Water, Solvent	3s apply
		Primer B: 03041101	Phosphoric acid monomer, Solvent, Dyes	Gently air-dry
		Paste: 02040901	TEGDMA, S-PRG filler, Bis-GMA	20s light-cure
Transbond XT (TB)	3M Unitek, Monrovia, California, USA	Transbond Plus self-etching primer: 204758	Methacrylated phosphoric acid esters, Amino benzoate, Camphorquinone	3s apply Gently air-dry
		Paste: 5MU	Bis-GMA, TEGDMA, Silane-treated quartz, Amorphous silica, Camphorquinone	20s light-cure
Kurasper F (KF)	Kuraray Medical, Tokyo, Japan	K-etchant: 00353B	37% Phosphoric acid	40s apply
		F-bond: 00036B	TEGDMA, 2-HEMA, Bis-GMA, Methyl methacrylate-methacryloyl fluoride copolymer, Sodium fluoride, Silanated silica filler, Initiators	20s wash Strongly air-dry
		Paste: 00026F	TEGMA, Bis-GMA, Silanated glass filler, Initiators	40s light-cure
Orthomite Superbond (OS)	Sun Medical, Moriyama, Japan	Red Activator: LE4	65% Phosphoric acid	30s apply
		Powder: KX2	PMMA	20s wash
		Liquid: LF3	MMA, 4-META	Strongly air-dry
		Catalyst: LE61	Tri-n-butylborane	Chemical-cure

TEGDMA: triethylene glycol dimethacrylate; PRG: pre-reacted glass ionomer; Bis-GMA: bisphenol A diglycidyl ether dimethacrylate; HEMA: 2-hydroxyethyl methacrylate; MMA: methyl methacrylate; PMMA: polymethyl methacrylate; 4-META: 4-methacryloyloxyethyl trimellitate anhydride

adhesive paste includes S-PRG (surface pre-reacted glass ionomer) filler particles, which are formed by an acid-base reaction between fluoroaluminosilicate glass and a polyalkenoic acid in the presence of water<sup>12</sup>. S-PRG fillers can release and recharge fluoride ions<sup>12</sup>.

Transbond XT is composed of a fluoride-free light-cured adhesive system. Transbond Plus self-etching primer was used as a conditioner, which contains methacrylated phosphoric acid esters.

Kurasper F is composed of a phosphoric acid etchant and a fluoride-releasing light-cured adhesive system in which sodium fluoride is the source of fluoride ion release.

Orthomite Superbond is composed of a phosphoric acid etchant and a fluoride-free chemically cured adhesive system which consists of PMMA powder, a

liquid component (MMA and 4-META), and a chemical initiator (tri-n-butylborane)<sup>13,14</sup>.

#### *Specimen preparation*

Eighty freshly extracted bovine incisors free of obvious defects were stored frozen prior to use. The roots of the teeth were cut off, leaving the crowns, which were embedded in a chemically cured acrylic resin (Unifast Trad, GC, Tokyo, Japan) in an acrylic tube to allow for standardized and secure placement during testing. The facial enamel surface was parallel to and about 1 mm above the cylinder rim. Then, the exposed enamel was flattened with 600-grit silicone carbide paper under copious water to provide an area for bonding. The enamel surfaces were then cleaned ultrasonically in distilled water. After which, the specimens were randomly divided into

eight groups.

Orthodontic metal brackets (One Piece Bracket, Kanno, Nagareyama, Japan) with a bonding area of 16.96 mm<sup>2</sup> were bonded to the enamel surface according to the manufacturers' instructions (Table 1).

All bonding procedures were performed by the same operator. Excess adhesive was carefully removed, and the BO, TB, and KF specimens were light-cured with a visible light curing unit (Optilux 500, Sybron Kerr Corp., USA), while the specimens of OS were chemically cured at room temperature. For BO and TB, the specimens were light-cured for 20 seconds (10 seconds from the mesial edge and 10 seconds from the distal edge of bracket). For KF, the specimens were light-cured for 40 seconds (20 seconds from mesial and 20 seconds from distal).

#### Shear bond test

Prepared specimens were left at room temperature for 30 minutes, and then stored in one of the two conditions as follows: deionized water at 37 °C for 24 hours (TC-0) or deionized water at 37 °C for 24 hours followed by thermal cycling of 5000 times (5-55 °C, dwell time of 30 seconds each) (TC-5000). Thermal cycling is a well-known *in vitro* durability test, which accelerates water penetration through the interface between the adhesive and enamel. Bishara *et al.*<sup>15</sup> evaluated the bonding durability of orthodontic brackets using thermal cycling of 500 times. Sirirungrojying *et al.*<sup>10</sup> also evaluated bonding durability using thermal cycling of 2000 and 5000 times. In this study, thermal cycling was determined at 5000 times.

Shear bond test was performed for each specimen

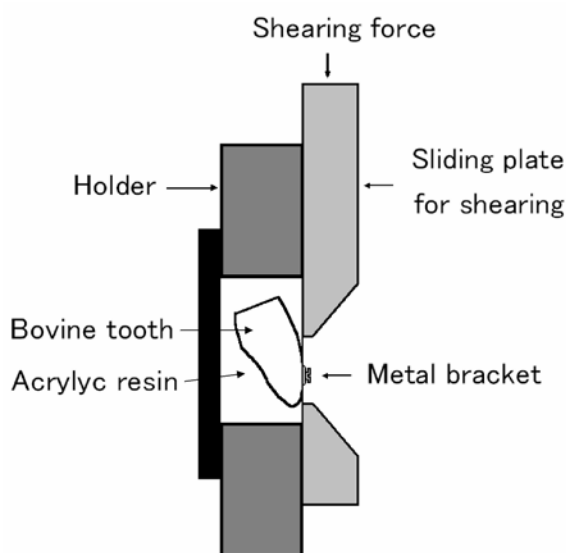


Fig. 1 Schematic drawing of shear bond strength measurement.

using an ISO standard jig<sup>16</sup> in the same manner as described by Ikeda *et al.*<sup>17</sup>. A universal testing machine (AG 500B, Shimadzu, Kyoto, Japan) was used for the shear bond test at a crosshead speed of 1 mm/min as shown in Fig. 1. Each tooth was oriented so that its facial surface was parallel to the direction of force during the shear bond test. The force was directly applied to the bracket-tooth interface. Load at bracket failure was recorded by a personal computer connected to the testing machine. Shear bond strength value was calculated in MPa by dividing the force by the area of the bracket base.

#### Failure mode analysis

After debonding, the teeth and brackets were examined under  $\times 10$  magnification with an optical microscope (OME-GWHIO, Olympus Co., Tokyo, Japan) and scored according to the criteria of the adhesive remnant index (ARI)<sup>18</sup> as follows: 0 - No adhesive left on tooth; 1 - Less than half of the adhesive left on tooth; 2 - More than half of the adhesive left on tooth; 3 - All the adhesive left on the tooth, with distinct impression of the bracket mesh.

#### SEM observation

The ground enamel surface and enamel surfaces conditioned with each adhesive system were observed with a SEM. The enamel surface was ground with 600-grit silicone carbide paper under copious water and ultrasonically cleaned in distilled water. For BO and TB, the self-etching primer of each system was applied to the ground enamel surface according to the manufacturers' instructions, gently air-blown, rinsed with acetone for 30 seconds, and then rinsed with distilled water for 30 seconds followed by gentle air-drying. For KF and OS, the enamel surface was etched with phosphoric acid, rinsed with water for 30 seconds, and gently air-dried.

To observe the debonded surface of the specimens, representative debonded specimens were selected from each group. All specimens were gold sputter-coated before examination with a SEM (JSM 5310LV, JOEL, Tokyo, Japan).

#### Statistical analysis

The number of specimens per group for shear bond testing was 10. Shear bond strengths were analyzed by two-way analysis of variance (ANOVA) using the Statistical Package for Medical Science (SPSS Ver.11 for Windows) for statistical procedures. The factors analyzed were material and storage condition. Following this, Tukey's HSD test was performed. ARI scores of the mode of failure were analyzed using Steel-Dwass nonparametric multiple comparison test by Tukey's procedure. The statistical calculations of mode of failure were performed using a statistical software, KyPlot (Version 3.0 for

Windows, Keyence Incorporated, Tokyo, Japan). Significance for all statistical tests was predetermined at a 95% confidence level.

RESULTS

*Shear bond strength*

The shear bond strengths of the four orthodontic adhesives to enamel are summarized in Table 2. Statistical significance of the bond strengths to enamel is shown in Table 3. Two-way ANOVA revealed that bond strength was influenced by both material (F=4.688, p=0.005) and storage condition (F=21.720, p=0.0001). There was also a significant interaction between the independent variables, material and storage condition (F=3.272, p=0.026).

In the TC-0 groups, there were no significant differences between BO and phosphoric acid-etching adhesive systems (KF and OS), while the bond strength of TB was significantly lower than those of

Table 2 Shear bond strengths to enamel (MPa).

	TC-0	TC-5000
BO	20.3 ± 4.7	18.8 ± 4.1
TB	17.6 ± 4.0	16.4 ± 3.3
KF	23.9 ± 3.8	17.5 ± 4.9
OS	24.9 ± 3.2	17.7 ± 2.8

n=10, Mean ±SD TC-0: Not thermocycled  
TC-5000: Thermocycled

phosphoric acid-etching adhesive systems (KF and OS). There were no significant differences between BO and TB, and between KF and OS, respectively (p>0.05).

In the cases of BO and TB, there were no significant differences in bond strength between TC-0 and TC-5000 (p>0.05). However, the bond strengths of KF

Table 3 Summary of the statistical analysis of the bond strengths to enamel using ANOVA supplemented with Tukey's HSD test.

Adhesive	TC	BO		TB		KF		OS	
		TC-0	TC-5000	TC-0	TC-5000	TC-0	TC-5000	TC-0	TC-5000
BO	TC-0								
	TC-5000	n.s.							
TB	TC-0	n.s.	n.s.						
	TC-5000	n.s.	n.s.	n.s.					
KF	TC-0	n.s.	n.s.	*	*				
	TC-5000	n.s.	n.s.	n.s.	n.s.	*			
OS	TC-0	n.s.	*	*	*	n.s.	*		
	TC-5000	n.s.	n.s.	n.s.	n.s.	*	n.s.	*	

Table 4 Frequency distribution of adhesive remnant index (ARI) scores.

	TC-0				TC-5000			
	ARI scores				ARI scores			
	0	1	2	3	0	1	2	3
BO	3	7	0	0	2	5	1	2
TB	6	3	1	0	4	5	1	0
KF	8	2	0	0	7	3	0	0
OS	1	2	2	5	2	1	2	5

ARI indicates adhesive remnant index; 0: No adhesive left on the tooth; 1: Less than half of the adhesive left on the tooth; 2: More than half of the adhesive left on the tooth; 3: All the adhesive left on the tooth with distinct impression of the bracket mesh.

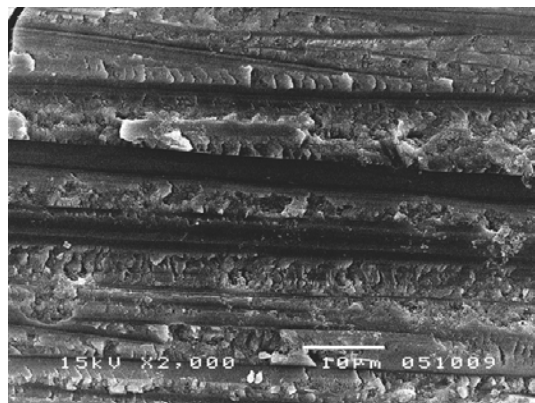


Fig. 2 SEM photograph of enamel surface ground with 600-grit silicone carbide paper ( $\times 2000$ ). Smear layer and scratch lines were observed.

and OS significantly decreased after 5000 times of thermal cycling ( $p < 0.05$ ).

In the TC-5000 groups, there were no significant differences in bond strength among the adhesive materials ( $p > 0.05$ ).

#### Modes of failure

The modes of failure according to the ARI index are summarized in Table 4. For each adhesive, no significant differences were observed between the TC-0 and TC-5000 groups ( $p > 0.05$ ). However, there were significant differences in the mode of failure between KF (TC-0) and OS (TC-0), and between KF (TC-5000) and OS (TC-0) ( $p < 0.05$ ). The predominant modes of bracket failure for BO, TB and KF were at the enamel-resin interface with less than 50% of the adhesive on the enamel surface, whereas the bracket-resin interface was the most common site of failure in OS.

#### SEM observation

Figure 2 shows an enamel surface ground with 600-grit silicone carbide paper. A smear layer was created on the ground surface with some scratch lines. On the other hand, the enamel surfaces conditioned with each adhesive are shown in Figs. 3(a) to (d).

The smear layer on the ground enamel was completely removed after conditioning by all the adhesive systems. However, the etched enamel patterns were different between the self-etching primers (Figs. 3(a) and (b)) and phosphoric acid etchants (Figs. 3(c) and 3(d)).

In the cases of BO and TB (Figs. 3(a) and (b)), enamel prisms were less prominent and so were the micro-irregularities of the hydroxyapatite crystals. In other words, the enamel surfaces were slightly roughened by the acidic primers. For KF and OS (Figs. 3(c) and (d)), the prismatic structure of the

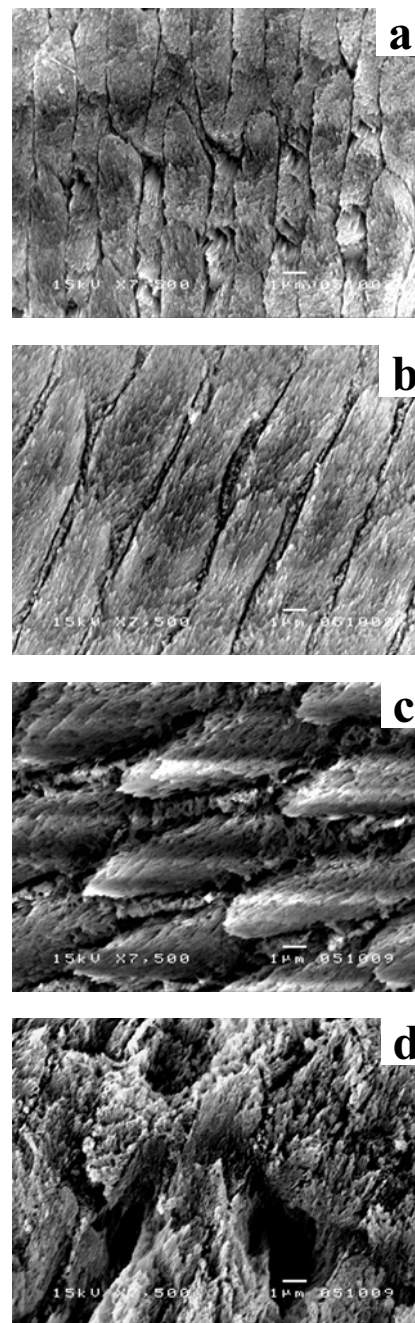


Fig. 3 SEM photographs of enamel surfaces conditioned with each adhesive ( $\times 7500$ ). a: Beauty Ortho Bond primer; b: Transbond Plus self-etching primer; c: 37% phosphoric acid in Kurasper F; d: 65% phosphoric acid in Orthomite Superbond. Enamel surfaces were slightly roughened by the acidic primer (a, b), while enamel surfaces were strongly etched and the prismatic structure of the enamel surface was selectively etched and easily identified (c, d).

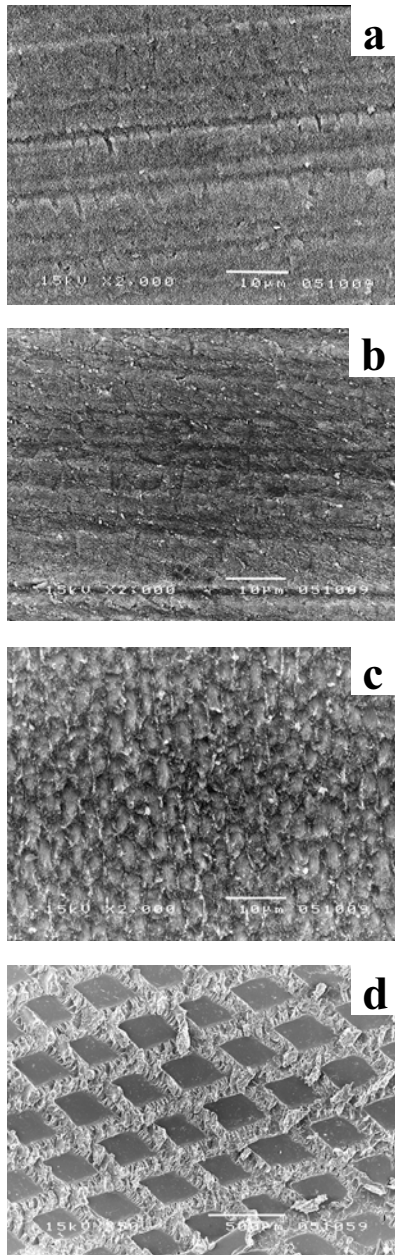


Fig. 4 SEM photographs of typical fracture surfaces on the enamel side after shear bond testing. a: Beauty Orthobond (TC-0) ( $\times 2000$ ); b: Transbond XT (TC-0) ( $\times 2000$ ); c: Kurasper F (TC-0) ( $\times 2000$ ); d: Orthomite Superbond (TC-0) ( $\times 50$ ). Partial adhesive failure was observed, and remnants of adhesive remained on the enamel surface (a, b, c). Distinct impression of the bracket mesh was observed (d). Mode of fracture tendency of each adhesive was not different between the TC-0 and TC-5000 groups.

enamel surface was selectively etched and easily identified. Micro-irregularity of the hydroxyapatite crystals was apparent.

Typical fracture surfaces on the enamel side after shear bond testing are depicted in Fig. 4. Partial adhesive failure occurred predominantly in the BO, TB and KF groups, in which remnants of the adhesives remained on the enamel surface (Figs. 4(a) to (c)). In the OS group, failures mainly occurred at the interface between the bracket and the adhesive in both TC-0 and TC-5000 conditions (Fig. 4(d)). Distinct impressions of the bracket mesh were observed.

## DISCUSSION

Direct bonding of orthodontic brackets using the acid-etch technique has become a common technique in the orthodontic field. Phosphoric acid etching produces a roughened enamel surface by dissolving calcium components and forming enamel resin tags. Although enamel etching technique is a useful and accepted procedure for bonding orthodontic brackets, there is a need to maintain clinically useful bond strengths while minimizing the amount of enamel loss.

Recent studies in operative dentistry have suggested that self-etching primers with lower decalcifying ability are less effective than phosphoric acid etching when used to bond to ground enamel or intact enamel<sup>19</sup>. Previously, Bishara *et al.*<sup>11</sup> found that the shear bond strength of orthodontic brackets after Prompt L-Pop self-etching primer treatment was significantly lower than that after phosphoric acid etching with TB. Yamada *et al.*<sup>20</sup> also found that the shear bond strength after SE Bond self-etching primer (Kurary Medical) treatment was significantly lower than that after phosphoric acid etching with KF. Similarly, the present study demonstrated that the shear bond strengths of brackets bonded with the self-etching adhesive systems (BO and TB) were lower than those bonded with phosphoric acid-etching adhesive systems (KF and OS) in the control groups. However, a bond strength of approximately 17 MPa was maintained after thermal cycling in the self-etching adhesive groups, while the bond strength significantly decreased after thermal stress in the phosphoric acid-etching groups. It should be pointed out that Martin and Garcia-Godoy<sup>21</sup> commented that high shear bond strength in orthodontics is not necessarily a beneficial property of an orthodontic adhesive, because enamel can be lost during the debonding procedure as well as during the removal of residual resin. Bishara *et al.*<sup>22</sup> reported that a shear bond strength of 7 MPa to the enamel was clinically acceptable for bonding to the enamel surface in orthodontic treatment. Data obtained in this study thus

suggested that all the adhesive systems evaluated were acceptable for routine clinical use.

The effects of thermal changes on the bond strength of resin-based materials to hard dental tissues, as well as on their mechanical properties (*i.e.*, fracture toughness, elastic modulus), have been well documented<sup>23,24</sup>. Thermal cycling stresses the bond between resin and tooth substance and might affect bond strength<sup>25</sup>. Christensen<sup>26</sup> commented that impressive *in vitro* bond strengths were transient when subjected to temperature changes in the mouth.

In the case of self-etching adhesive systems, the acidic monomers in the self-etching primers are responsible for both etching and bonding. As such, the depth of demineralized enamel corresponds to the depth of penetration of the adhesive to be polymerized. This mechanism thus circumvents problems associated with insufficient penetration as well as improves the quality of hybridization<sup>27-29</sup>, thereby ensuring an excellent mechanical lock.

Phosphoric acid etching creates resin tags for mechanical retention between enamel and resin<sup>30</sup>. However, the resin may not completely infiltrate etched enamel<sup>31</sup>. A region of unprotected enamel prisms might be susceptible to hydrolytic degradation after thermal cycling. In addition, water diffusion into the bonded interface between adhesive and tooth surface was found to cause the resin to swell and become plasticized<sup>32</sup>, as well as reduce enamel hardness due to loss of surface calcium<sup>33</sup>.

Basically, there are two opinions on the remaining adhesive following bracket debonding. One opinion largely favors bracket-adhesive interface failure with the adhesive resin left mainly on the enamel surface<sup>5,34</sup>, when a heavy-filled resin is used to bond orthodontic attachments. The microporosities created by etching are filled with the resin and provide mechanical retention. The second opinion favors failure at the enamel-adhesive resin interface, because there is less adhesive left to remove from the enamel surface after debonding<sup>34</sup>. Martin and Garcia-Godoy<sup>21</sup> suggested that a weaker adhesive with a lower strength value might be preferable so as to increase failure rate at the enamel-adhesive resin interface. In this way, minimal clean-up would be needed with reduced likelihood of damage to the enamel.

The predominant mode of bracket failure for BO, TB, and KF was at the enamel-resin interface with less than 50% of the adhesive on the enamel surface. In the cases of BO and TB, scratch lines with 600-grit silicone carbide paper were observed after debonding, which indicated that the self-etching adhesive systems did not damage the enamel surface during debonding.

As for OS, the predominant mode of bracket failure was at the bracket-adhesive interface with more than 50% of the adhesive on the enamel surface. OS

is MMA-based, the mechanical properties of which are weaker than the light-cured adhesive resins<sup>35,36</sup>. Hotta *et al.*<sup>37</sup> reported that 4-MET, a hydrolysis product of 4-META, promoted effective diffusion of monomers into enamel. SEM observations of the conditioned enamel surfaces revealed that the self-etching primers produced less enamel dissolution compared with phosphoric acid etching. Moreover, the morphological appearances of the enamel surface were different between KF and OS, which was probably due to the different concentrations of phosphoric acid in the etchants. The 37% phosphoric acid of K-etchant in KF was more aggressive than the 65% solution of Red Activator in OS<sup>31</sup>.

Decalcification is a common side effect of fixed appliance orthodontic treatment<sup>38</sup>. On this note, orthodontic treatment with multibonded appliances poses a significant caries risk<sup>6,39</sup>. To counter this problem, fluoride-releasing composites for bonding brackets have attracted considerable attention and garnered much interest. This is because they may inhibit the decalcification of enamel around the brackets by delivering fluoride to the affected environment<sup>40,41</sup>. Furthermore, the remineralization capability and antibacterial property of fluoride may help eliminate the risk of dental caries<sup>42</sup>. BO contains S-PRG filler for the release and uptake of fluoride ions, and might thus prevent demineralization but facilitate remineralization of the surrounding enamel<sup>6</sup>.

Orthodontic bracket bonding is performed on intact enamel. The intact enamel surface is hypermineralized and contains more fluoride than ground enamel. Prismless layer on enamel surface<sup>43</sup> is less conducive to bonding by conventional acid gel conditioning<sup>44</sup> and self-etching primer application<sup>45</sup>. Kanemura *et al.*<sup>45</sup> reported that bond strength was significantly reduced when self-etching systems were bonded to intact enamel. Similarly, Senawongse *et al.*<sup>46</sup> reported that self-etching adhesive systems exhibited significantly lower bond strengths than the phosphoric acid-etching adhesive systems on intact enamel. However, no statistically significant differences were found between self-etching adhesive systems and phosphoric acid-etching adhesive systems to ground enamel. Furthermore, bond strengths of self-etching adhesive systems to ground enamel were significantly higher than those to unground enamel, whereas phosphoric acid etching systems showed no such significant differences between intact and ground enamel. Based on the results obtained to date, further research is indeed needed to clarify whether self-etching primer adhesive systems could provide sufficient bond strength to intact human enamel.

## CONCLUSIONS

Self-etching adhesives, Beauty Ortho Bond and Transbond XT, showed more stable bond strengths to ground enamel after thermal cycling than the phosphoric acid-etching adhesives, Kurasper F and Orthomite Superbond. In addition, with self-etching adhesives, problems concerning the decalcification of and damage to the enamel surface were eliminated.

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## REFERENCES

- 1) Buonocore MG. A simple method of increasing the adhesion of acrylic filling materials to enamel surfaces. *J Dent Res* 1955; 34:849-853.
- 2) Zarrinnia K, Eid NM, Kehoe MJ. The effect of different debonding techniques on the enamel surface: an *in vitro* qualitative study. *Am J Orthod Dentofacial Orthop* 1995; 108:284-293.
- 3) Thanos CE, Munholland T, Caputo AA. Adhesion of mesh base direct bonding brackets. *Am J Orthod* 1979; 75:421-430.
- 4) Gorelick L. Bonding metal brackets with a self-polymerizing sealant-composite: a 12-month assessment. *Am J Orthod Dentofacial Orthop* 1977; 71:542-553.
- 5) Proffit W. Contemporary orthodontics, CV Mosby Co, St Louis, 1986, pp.282-295.
- 6) O'Reilly MM, Featherstone JD. Demineralization and remineralization around orthodontic appliances: an *in vivo* study. *Am J Orthod Dentofacial Orthop* 1987; 92:33-40.
- 7) Chigira H, Koike T, Hasegawa T, Itoh K, Wakumoto S, Hayakawa T. Effect of the self etching dentin primers on the bonding efficacy of a dentin adhesive. *Dent Mater J* 1989; 8:86-92.
- 8) Han L, Cruz E, Fukushima M, Okamoto A, Okiji T. An ultrastructural analysis of the prototype single-step adhesive applied on enamel and dentin surfaces. *Dent Mater J* 2004; 23:321-328.
- 9) Cacciafesta V, Sfondrini MF, De Angelis M, Scribante A, Klersy C. Effect of water and saliva contamination on shear bond strength of brackets bonded with conventional, hydrophilic, and self-etching primers. *Am J Orthod Dentofacial Orthop* 2003; 123:633-640.
- 10) Sirirungrojying S, Hayakawa T, Saito K, Meguro D, Nemoto K, Kasai K. Bonding durability between orthodontic brackets and human enamel treated with megabond self-etching primer using 4-META/MMA-TBB resin cement. *Dent Mater J* 2004; 23:251-257.
- 11) Bishara SE, Von Wald L, Laffoon JF, Warren JJ. Effect of a self-etch primer/adhesive on the shear bond strength of orthodontic brackets. *Am J Orthod Dentofacial Orthop* 2001; 119:621-624.
- 12) Ikemura K, Tay FR, Kouro Y, Endo T, Yoshiyama M, Miyai K, Pashley DH. Optimizing filler content in an adhesive system containing pre-reacted glass-ionomer fillers. *Dent Mater* 2003; 19:137-146.
- 13) Kameyama A, Ihara S, Amagai T, Miake Y, Kawada E, Oda Y, Yanagisawa T, Hirai Y. Adhesion of 4-META/MMA-TBB resin to heated dentin: effects of pre-treatments with FeCl<sub>3</sub> and/or HEMA. *Dent Mater J* 2003; 22:1-9.
- 14) Hirabayashi C. Studies on MMA-TBB resin II. The effect of dual use of TBB and other initiators on polymerization of PMMA/MMA resin. *Dent Mater J* 2003; 22:48-55.
- 15) Bishara SE, Ajlouni R, Laffoon JF. Effect of thermocycling on the shear bond strength of a cyanoacrylate orthodontic adhesive. *Am J Orthod Dentofacial Orthop* 2003; 123:21-24.
- 16) Noguchi H, Nakano M, Kumasaka A, Etchu Y. Evaluation of Japanese dental adhesives. *Dent Eng* 1982; 60:16-27.
- 17) Ikeda M, Nikaido T, Foxton RM, Tagami J. Shear bond strengths of indirect resin composites to hybrid ceramic. *Dent Mater J* 2005; 24:238-243.
- 18) Artun J, Bergland S. Clinical trials with crystal growth conditioning as an alternative to acid-etch enamel pretreatment. *Am J Orthod* 1984; 85:333-340.
- 19) Pashley DH, Tay FR. Aggressiveness of contemporary self-etching adhesives. Part II: Etching effects on unground enamel. *Dent Mater* 2001; 17:430-444.
- 20) Yamada R, Hayakawa T, Kasai K. Effect of using self-etching primer for bonding orthodontic brackets. *Angle Orthod* 2002; 72:558-564.
- 21) Martin S, Garcia-Godoy F. Shear bond strength of orthodontic brackets cemented with a zinc oxide-polyvinyl cement. *Am J Orthod Dentofacial Orthop* 1994; 106:615-620.
- 22) Bishara SE, Gordan VV, Von Wald L, Jakobsen JR. Shear bond strength of composite, glass ionomer, and acidic primer adhesive systems. *Am J Orthod Dentofacial Orthop* 1999; 115:24-28.
- 23) Miyazaki M, Sato M, Onose H. Durability of enamel bond strength of simplified bonding systems. *Oper Dent* 2000; 25:75-80.
- 24) Price RB, Derand T, Andreou P, Murphy D. The effect of two configuration factors, time, and thermal cycling on resin to dentin bond strengths. *Biomaterials* 2003; 24:1013-1021.
- 25) Nikaido T, Kunzelmann KH, Chen H, Ogata M, Harada N, Yamaguchi S, Cox CF, Hickel R, Tagami J. Evaluation of thermal cycling and mechanical loading on bond strength of a self-etching primer system to dentin. *Dent Mater* 2002; 18:269-275.
- 26) Christensen GJ. Has tooth structure been replaced? *J Am Dent Assoc* 2002; 133:103-105.
- 27) Weerasinghe DS, Nikaido T, Wettasinghe KA, Abayakoon JB, Tagami J. Micro-shear bond strength and morphological analysis of a self-etching primer adhesive system to fluorosed enamel. *J Dent* 2005; 33:419-426.
- 28) Nakabayashi N, Pashley DH. Hybridization of dental hard tissue, Quintessence Publishing Co, Tokyo, 1998, pp.6-7.
- 29) Gordan VV, Vargas MA, Cobb DS. Evaluation of adhesive systems using acidic primers. *Am J Dent* 1997; 10:219-223.
- 30) Soetopo, Beech DR, Hardwick JL. Mechanism of adhesion of polymers to acid-etched enamel. Effect of acid concentration and washing on bond strength. *J*



- Oral Rehabil 1978; 5:69-80.
- 31) Shinchi MJ, Soma K, Nakabayashi N. The effect of phosphoric acid concentration on resin tag length and bond strength of a photo-cured resin to acid-etched enamel. *Dent Mater* 2000; 16:324-329.
- 32) Soderholm KJ. Correlation of *in vivo* and *in vitro* performance of adhesive restorative materials: a report of the ASC MD156 Task Group on Test Methods for the Adhesion of Restorative Materials. *Dent Mater* 1991; 7:74-83.
- 33) Muhlemann HR. Storage medium and enamel hardness. *Helv Odontol Acta* 1964; 8:112-117.
- 34) Bishara SE, Ajlouni R, Laffoon JF. Comparison of the shear bond strength of 2 self-etch primer/adhesive systems. *Am J Orthod Dentofacial Orthop* 2004; 125:348-350.
- 35) Burrow MF, Tagami J, Negishi T, Nikaido T, Hosoda H. Early tensile bond strengths of several enamel and dentin bonding systems. *J Dent Res* 1994; 73:522-528.
- 36) Kitasako Y, Burrow MF, Nikaido T, Tagami J. Long-term tensile bond durability of two different 4-META containing resin cements to dentin. *Dent Mater* 2002; 18:276-280.
- 37) Hotta K, Mogi M, Miura F, Nakabayashi N. Effect of 4-MET on bond strength and penetration of monomers into enamel. *Dent Mater* 1992; 8:173-175.
- 38) Millett DT, Nunn JH, Welburry RR, Gordon PH. Decalcification in relation to brackets bonded with glass ionomer cement or a resin adhesive. *Angle Orthod* 1999; 69:65-70.
- 39) Ogaard B. Prevalence of white spot lesions in 19-year-olds: a study on untreated and orthodontically treated persons 5 years after treatment. *Am J Orthod Dentofacial Orthop* 1989; 96:423-427.
- 40) Mitchell L. An investigation into the effect of a fluoride releasing adhesive on the prevalence of enamel surface changes associated with directly bonded orthodontic attachments. *Br J Orthod* 1992; 19:207-214.
- 41) Cildir SK, Sandalli N. Fluoride release/uptake of glass-ionomer cements and polyacid-modified composite resins. *Dent Mater J* 2005; 24:92-97.
- 42) Imazato S. Antibacterial properties of resin composites and dentin bonding systems. *Dent Mater* 2003; 19:449-457.
- 43) Whittaker DK. Structural variations in the surface zone of human tooth enamel observed by scanning electron microscopy. *Arch Oral Biol* 1982; 27:383-392.
- 44) Nathanson D, Bodkin JL, Evans JR. SEM of etching patterns in surface and subsurface enamel. *J Pedodont* 1982; 7:11-17.
- 45) Kanemura N, Sano H, Tagami J. Tensile bond strength to and SEM evaluation of ground and intact enamel surfaces. *J Dent* 1999; 27:523-530.
- 46) Senawongse P, Sattabanasuk V, Shimada Y, Otsuki M, Tagami J. Bond strengths of current adhesive systems on intact and ground enamel. *J Esthet Restor Dent* 2004; 16:107-115.