Enamel bonding of self-etching and phosphoric acid-etching orthodontic adhesives in simulated clinical conditions: Debonding force and enamel surface

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This study aimed to evaluate the effectiveness of self-etching and phosphoric acid-etching orthodontic adhesives for enamel bonding in simulated clinical conditions. By using two self-etching (Transbond Plus, TP; Beauty Ortho Bond, BB) and two acid-etching (Transbond XT, TX; Superbond Orthomite, SB) adhesives, orthodontic brackets were bonded on human premolars (n=10 for each adhesive). Ten teeth without bracket bonding, i.e., intact enamel surfaces, were used as control for SEM observation. After 7-day storage in lactic acid solution, bracket debonding force by means of debonding pliers, adhesive remnant index (ARI), and enamel surface morphology were examined. All the tested adhesives exhibited sufficient bond strength for clinical use. The ARI scores were almost the same among the four adhesives. In terms of SEM observation, the enamel surfaces in the control and TP groups showed a slight change after immersion in lactic acid solution, while the BB group showed less change on the enamel surface compared with the TP group. Meanwhile, the two acid-etching adhesives caused considerable demineralization. Taken together, these findings indicated that the action of self-etching systems was evidently more conservative.

Keywords: Self-etching, Phosphoric acid-etching, Orthodontic adhesive

INTRODUCTION

Adhesives for orthodontic brackets, such as composite resins, have been considered as one of the most significant developments in clinical orthodontics. Nowadays, the use of acid-etching systems when attaching brackets to the enamel surface with orthodontic adhesives has been widely accepted by most orthodontists as a routine technique. This bonding system provides a strong adhesion of the brackets to the tooth surface, hence preventing bracket failures. However, acid-etching produces iatrogenic effects on the enamel surface, especially during bracket removal. Reports of enamel fractures and cracks when debonding raised questions about the safety of the various procedures used to remove these attachments.

Recently, self-etching systems have been introduced by the market as a substitute for phosphoric acid-etching systems. These self-etching systems combine the etching and priming procedures into one step and eliminate the need for separate etching, rinsing, and drying steps. Owing to these features, self-etching systems are also known as bicomponent hydrophilic adhesives. When compared with phosphoric acid etching, self-etching systems demonstrate a more conservative etch pattern, a smaller amount of demineralization, and less adhesive penetration of the enamel surface. Currently, when it comes to bracket bonding to the enamel surface, self-etching systems have emerged as a favorite alternative to the separate two-step etching and primer system.

Enamel demineralization occurs frequently around and beneath orthodontic brackets. This is probably because fixed orthodontic appliances create extra retention sites, hence harboring more mutans streptococci and leading to increased risk for caries development. Another factor that is known to influence the processes of carious lesion formation is the ambient level of fluoride, and typical subsurface caries-like lesions were induced on the enamel surface in the absence of fluoride. In other words, fluoride can reduce the severity of subsurface caries lesions, which means that fluoride-releasing orthodontic adhesives may inhibit caries development on tooth surfaces adjacent to fixed orthodontic appliances.

In this study, we observed the enamel surfaces treated with two self-etching and two phosphoric acid-etching systems containing different amounts of fluoride after 7-day immersion in the lactic acid solution. The aim of this study was to evaluate the effectiveness of self-etching and phosphoric acid-etching orthodontic adhesives for enamel bonding in simulated clinical conditions by means of the following variables: the magnitude of force required to debond the bracket, the amount of residual adhesive remaining on the enamel surface, and the enamel surface morphology after debonding.
MATERIALS AND METHODS

Tooth specimens
A total of 50 extracted human premolars were obtained and stored in thymol solution (0.1% wt/vol). All acquired teeth were extracted for orthodontic treatment, and informed consent was obtained from each subject prior to the experiments. The tooth selection criterion was absence of large restorations or caries that may affect enamel strength and morphology. Prior to the experiments, all tooth specimens were cleaned and polished with a fluoride-free paste (Pressage, Shofu Inc., Kyoto, Japan) using a low-speed handpiece (10 seconds). They were then thoroughly washed with water and air-dried immediately before the bonding procedure.

Adhesive systems
Materials used in this study are listed in Table 1. Four orthodontic adhesive systems (Transbond Plus and Transbond XT, 3M Unitek, Monrovia, CA, USA; Beauty Ortho Bond, Shofu Inc., Kyoto, Japan; Superbond Orthomite, Sun Medical Co. Ltd., Shiga, Japan) were employed in this study. For each orthodontic adhesive, 10 tooth specimens were randomly selected and bonded with a stainless steel orthodontic bracket (0.018 inches; Standard Edge, Dyna-Lock, 3M Unitek, Seefeld, Germany) each. Any overflowed adhesive paste was removed immediately. Bonding procedure was then performed according to the instructions of each manufacturer as follows:

- Beauty Ortho Bond (BB group): Tooth specimens were conditioned with a self-etch primer after mixing two components. The adhesive paste contained 1.72% of fluoride. After surface treatment, the bracket was placed on the enamel surface, bonded with a fluoride-releasing paste, and light-cured (Coltolux 4, Coltene/Whaledent Inc., OH, USA) for 30 seconds. Light-curing was divided into three time lapses (10 seconds each) on the mesial, distal, and occlusal sides respectively.
- Transbond Plus (TP group): Tooth specimens were conditioned with a self-etching primer after mixing two components according to the manufacturer’s instructions. The adhesive paste contained 0.47% of fluoride. Brackets were bonded with Transbond XT and light-cured using the same protocol as that of Beauty Ortho Bond.
- Transbond XT (TX group): Tooth specimens were etched (35% H₃PO₄; 30 seconds), washed for 20 seconds with an air-water spray, and dried to a chalky white appearance. Then, the Transbond XT primer was applied to the etched surface. The paste contained 0.47% of fluoride. Brackets were bonded with Transbond XT and light-cured in the same manner as described above.
- Superbond Orthomite (SB group): Tooth specimens were etched (65% H₃PO₄; 30 seconds), washed, and dried using the same protocol as that of Transbond XT. Superbond Orthomite, 4-META/MMA-TBB resin, was a chemical-cure adhesive system.

### Table 1  Materials used in this study

<table>
<thead>
<tr>
<th>Material</th>
<th>Component</th>
<th>Composition</th>
<th>Manufacturer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beauty Ortho Bond (BB)</td>
<td>Primer A</td>
<td>Water, Solvent</td>
<td>Shofu, Kyoto, Japan</td>
</tr>
<tr>
<td></td>
<td>Primer B</td>
<td>Phosphoric acid monomer, solvent</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Paste</td>
<td>TEGDMA, S-PRG filler, Bis-GMA</td>
<td></td>
</tr>
<tr>
<td>Transbond Plus (TP)</td>
<td>Self-etching Primer</td>
<td>Methacrylated phosphoric acid esters, Aminobenzoate, Camphorquinone</td>
<td>3M Unitek, Monrovia, California, USA</td>
</tr>
<tr>
<td></td>
<td>Paste</td>
<td>TEGDMA, Bis-GMA, silane-treated quartz, amorphous silica, Camphorquinone</td>
<td></td>
</tr>
<tr>
<td>Transbond XT (TX)</td>
<td>Etching gel</td>
<td>35% Phosphoric acid</td>
<td>3M Unitek, Monrovia, California, USA</td>
</tr>
<tr>
<td></td>
<td>Primer</td>
<td>TEGDMA, Bis-GMA</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Paste</td>
<td>TEGDMA, Bis-GMA, silane-treated quartz, amorphous silica, Camphorquinone</td>
<td></td>
</tr>
<tr>
<td>Superbond Orthomite (SB)</td>
<td>Red activator</td>
<td>65% Phosphoric acid</td>
<td>Sun Medical, Moriyama, Japan</td>
</tr>
<tr>
<td></td>
<td>Polymer</td>
<td>PMMA</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Monomer</td>
<td>MMA, 4-META</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Catalyst</td>
<td>Tri-n-butylborane</td>
<td></td>
</tr>
</tbody>
</table>

TEGDMA: triethylene glycol dimethacrylate; PRG: pre-reacted glass ionomer; Bis-GMA: bisphenol A diglycidyl ether dimethacrylate; PMMA: polymethyl methacrylate; MMA: methyl methacrylate; 4-META: 4-methacryloyloxyethyl trimellitate anhydride
adhesive that comprised a mixture of a power and two liquids. The power and liquid components contained no fluoride. Brackets with then bonded with the mixed adhesive paste. The remaining 10 tooth specimens served as control, on which no etching and priming treatments were conducted nor any bracket placement. As for all the tooth specimens in each adhesive group, they were stored for 7 days in 0.1 M lactic acid solution (pH 4.0) at 37°C in an incubator after surface treatment and bracket placement.

Debonding strength measurement
A universal testing machine (AG100, Shimadzu Co., Kyoto, Japan) was used to measure the debonding strength (Fig. 1) at a crosshead speed of 5.0 mm/min. All the brackets were removed with a pair of debonding pliers (Ormco Co., CA, USA) according to the modified method of Bishara et al.3). The stainless steel blades of the pliers were placed at the bracket-adhesive interface, and a squeezing action was applied until bond failure occurred. The relationship between the measured failure load \( F_1 \) and the debonding strength \( F_2 \) is given as follows:

\[
F_1 M_1 = F_2 M_2
\]

Then,

\[
F_2 = F_1 M_1 / M_2
\]

where \( M_1 \) and \( M_2 \) are the moment arms in Fig. 1. The debonding strength \( F_2 \) in Newtons was divided by 14.1 mm² (cross-sectional area of the bracket bonding surface as provided by the manufacturer and confirmed by measurement) to determine the debonding strengths in MPa.

Adhesive remnant index (ARI)
All visible residual adhesive was carefully removed from the bonded surfaces with a pair of resin-removing pliers. Furthermore, similar to clinical use, the location of adhesive failure on the tooth surface was cleaned using a rubber cup with a fluoride pumice paste (Merssage, Shofu Inc., Kyoto, Japan) attached to a slow-speed handpiece for 10 seconds. After which, ARI assessment was performed.

Modified ARI scores in the range of 0 to 4 were determined as follows: 0 indicated that no adhesive remained on the tooth surface, implying that bond fracture occurred at the adhesive-enamel interface; 1 indicated that less than 25% of the adhesive remained on the tooth surface; 2 indicated that more than 25% but less than 50% of the adhesive remained on the tooth surface; 3 indicated that more than 50% but less than 75% of the adhesive remained on the tooth surface; 4 indicated that 75–100% of the adhesive remained on the tooth surface with a distinct impression of the bracket base, implying that most of the bond fracture occurred at the bracket-adhesive interface.

Enamel surface morphology
A scanning electron microscope (SEM; JSM-5300, Japan Electron Optics Laboratory Co. Ltd., Tokyo, Japan) was used to examine the enamel surfaces after debonding. The specimens were placed on aluminum stabs and coated with osmium for 10 seconds.

Statistical analysis
To evaluate the measured debonding strengths and ARI scores, means and standard deviations were calculated for each group (n=10). All the data were tested for normality of distribution (Kolmogorov–Smirnov test) and for uniformity (Bartlett’s test). Differences in the debonding strength and ARI score among the four adhesive groups were tested by one-way analysis of variance with a post-hoc test (Bonferroni test) for multiple comparisons. A probability of less than 0.05 for similarity of distribution was considered to be significantly different.

RESULTS
Debonding strength
Among the four adhesives, the highest debonding strength was achieved by SB at 7.6±0.7 MPa, followed by TX at 6.1±0.6 MPa (Fig. 2). TP and BB, which used self-etching primers, showed significantly (p<0.05) lower debonding strengths at 5.0±0.4 MPa and 4.6±0.7 MPa respectively, when compared to SB. Nonetheless, all the adhesives showed sufficient bond strength for clinical use.

ARI
Table 2 shows the ARI scores after the debonding procedure. The ARI score of SB after cleaning with fluoride pumice paste was 0.6±0.5, which was the highest, followed by TX and TP at 0.3±0.5 and 0.2±0.4
respectively. The ARI score of BB was the lowest at 0.1±0.3. However, no significant differences were found among the four adhesive groups.

**Enamel surface morphology**

Based on SEM observation, the enamel surface in TP group showed a slight change with shallow depressions when compared with that of the intact enamel surface (Figs. 3A and B). This was probably due to immersion in lactic acid solution. For the BB group, the etch pattern and shallow depressions were less uniform and in less areas than seen in the TP group (Fig. 3C). Meanwhile, TX and SB showed dissolution of the prism cores and peripheries and areas of incomplete demineralization (Figs. 3D and E). These images implied that the action of both self-etching systems seemed to be more conservative than phosphoric acid-etching.

After cleaning with fluoride pumice paste, the enamel surfaces beneath the brackets in TP and BB groups appeared to be considerably smooth (Figs. 4A and B). In contrast, the enamel surfaces in TX and SB groups appeared to be very rough with incomplete

<table>
<thead>
<tr>
<th>Group</th>
<th>ARI score</th>
</tr>
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<tbody>
<tr>
<td>TP</td>
<td>0.2 ± 0.4</td>
</tr>
<tr>
<td>BB</td>
<td>0.1 ± 0.3</td>
</tr>
<tr>
<td>TX</td>
<td>0.3 ± 0.5</td>
</tr>
<tr>
<td>SB</td>
<td>0.6 ± 0.5</td>
</tr>
</tbody>
</table>

Table 2  Adhesive remnant index (ARI) scores (mean ± SD) of four adhesive systems

Fig. 2  Debonding strengths of four adhesive systems. Error bars indicate a standard deviation. *: Significant difference at p<0.05, **: p<0.01.

Fig. 3  Scanning electron images of enamel surfaces surrounding the brackets after 7-day immersion in lactic acid solution. A: Untreated enamel (control); B: TP; C: BB; D: TX; E: SB. Scale bars=50 µm.
deminerlization, thereby implying an irreversible damage to the enamel due to acid-etching (Figs. 4C and D).

DISCUSSION

The efficacy of a self-etching system in terms of bond strength has been investigated in numerous studies\(^1,2,7,8,15-20\). Majority of the previous studies demonstrated that the shear bond strength of self-etching systems was significantly less than that of conventional acid-etching systems\(^1,2,8,15-18\), although a few studies indicated no significant differences in bond strength between self-etching and acid-etching systems\(^7,19,20\).

In this study, the debonding forces of two self-etching systems were significantly lower than that of the acid-etching system (SB group), a finding similar to those of previous studies. However, while the debonding strength measured by Bishara et al.’s method employed in this study was approximately 40% lower than conventional shear bond strength\(^21\), the mean bond strength of both self-etching systems was higher than the average suggested by Reynolds\(^22\) as a minimum for clinical orthodontic treatment. This meant that apart from acid-etching systems, self-etching systems provide adequate bonding strength for orthodontic treatment purposes. Indeed, in a study by Elekdag-Turk et al.\(^23\) to compare the clinical performance of self-etching and acid-etching systems over a 6-month period, they reported that the failure and survival rates of orthodontic brackets did not show significant differences between the two adhesive systems. Therefore, the findings of Elekdag-Turk et al.\(^23\) further corroborated with the results of the present study in that self-etching system can be effectively used for the bonding of orthodontic brackets.

On ARI scores, BB exhibited the lowest score after the debonding procedure but which was not significantly different from the other three adhesive groups. This meant that enamel clean-up after debonding should be easier and faster with self-etching systems, hence reduced chairside time too.

On enamel surface morphology, the SEM images of both self-etching systems showed a more conservative action than that of the acid-etching system. These findings were in agreement with previous studies\(^1,2,20\). Although the acid-etching treatment provides a strong adhesion of the brackets to the tooth surface, it may produce iatrogenic effects on the enamel surface by
which enamel cracks and fractures sometimes occur when debonding. Since the damage to the enamel surface still remained after mechanical polishing, this might lead to color alteration during and after orthodontic treatment. On the contrary, the enamel surfaces treated with self-etching primers were almost the same as that of the untreated tooth. In particular, the enamel surface treated with BB self-etching primer showed minimal change after immersion in lactic acid solution.

It is noteworthy that BB was filled with surface pre-reacted glass-ionomer (S-PRG) filler particles\(^2\text{-}^\text{28}\), which are known to be equipped with fluoride release and recharge abilities\(^2\text{-}^\text{23}\text{-}^\text{27}\). This implies that the enamel surface bonded with BB would be subjected to minimal change, as the fluoride release and recharge abilities of S-PRG filler would decrease enamel decalcification. Furthermore, glass-ionomers are equipped with the ability of neutralizing surrounding aqueous solutions, hence rendering the in vivo beneficial effect of inhibiting caries development\(^2\text{8}\). The latter mechanism would well complement that of fluoride release. However, surface enamel fluoride concentration was not measured in this study. Therefore, further investigations are necessary to verify the effect of fluoride release and recharge by the S-PRG filler.

**CONCLUSIONS**

Although the debonding strengths exhibited by both self-etching systems were significantly lower than those of acid-etching systems, these values were nonetheless higher than the average suggested as optimal for clinical use. Consistent with a lower debonding strength, the self-etching systems also exhibited a more conservative action than the acid-etching systems. In particular, the enamel surface bonded with a new fluoride-releasing adhesive (Beauty Ortho Bond) remained considerably smooth even after 7 days’ immersion in lactic acid solution. Enamel surface morphology findings of this study thus indicated that self-etching adhesive systems are well poised to be the effective means of eliminating enamel damage and/or decalcification during orthodontic treatment.

**REFERENCES**
