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Effects of long-term storage and thermocycling on bond strength of two self-etching primer adhesive systems

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SUMMARY The effects of 2 years of storage and 6000 thermocycles on the shear bond strength (SBS) of two self-etching adhesive systems were studied. Two self-etching primer (SEP) systems (Transbond Plus and Beauty Ortho Bond) and one etch and rinse system (Transbond XT) were used to bond brackets to 126 human premolars that were then stored in artificial saliva for 24 hours or 2 years and thermocycled in distilled water before SBS testing with a universal testing machine. The adhesive remnant index (ARI) scores were calculated. Data were compared by two-way analysis of variance and chi-square analysis. Enamel/adhesive interfaces were examined by scanning electron microscopy.

There was no significant difference in the mean SBS for the bonding materials among the three conditions. ARI scores showed that Transbond XT and Beauty Ortho Bond had less adhesive remaining on the teeth after ageing compared with storage for 24 hours. Specimens bonded with Beauty Ortho Bond showed leakage between the resin adhesive and enamel after ageing. Both SEP systems produced adequate SBS even after 2 years or 6000 times thermocycling. Thermocycling is an appropriate technique for determining the durability of orthodontic bracket bonding materials.

Introduction

After the concept of acid etching was introduced by Buonocore (1955), the direct bonding of orthodontic appliances to enamel with composite resin was introduced by Newman (1965) and is now widely accepted by most orthodontists (Eliades and Eliades, 2001; Eliades *et al.*, 2001). Bonding practices based on a self-etching primer (SEP), which combine etching and priming into a single step, are now being used in clinical orthodontics (Cehreli *et al.*, 2005; Arhun *et al.*, 2006; Bishara *et al.*, 2006; Faltermeier *et al.*, 2007; Scougall Vilchis *et al.*, 2007; Iijima *et al.*, 2008). In addition to saving time and reducing procedural errors, their lower etching ability, due to their higher pH compared with phosphoric acid, might minimize the potential for iatrogenic damage to enamel (Pashley and Tay, 2001; Zeppieri *et al.*, 2003).

Bracket-bonding failure sometimes occurs during the later stages of treatment due to heavy forces produced by an archwire or occlusal force. Bracket bond failure is not only frustrating for the practitioner but can also significantly affect treatment efficiency and have an economic impact on a practice (Northrup *et al.*, 2007). Although it has been demonstrated that the shear bond strengths (SBSs) of brackets bonded with SEP adhesive systems were similar to those with a conventional etch and rinse adhesive system (Scougall Vilchis *et al.*, 2007; Iijima *et al.*, 2008), most of these bonding studies measured short-term adhesive bond strength and did not extend the study period to encompass the duration of normal orthodontic treatment. Recently, Oesterle and Shellhart (2008) studied the effect of composite ageing for two conventional etch and rinse adhesive systems on SBS during a normal 24 month orthodontic treatment period and concluded that the SBS of orthodontic brackets increases from 30 minutes to 24 hours and then tends to decrease over the next 24 months.

The most commonly used artificial ageing technique is long-term water storage. Another widely used ageing technique is thermocycling. The International Organization for Standardization (ISO) TR 11450 standard (1994) indicates that a thermocycling regimen comprising 500 cycles in water between 5 and 55°C is an appropriate artificial ageing test, and many studies have been carried out following the ISO standard. However, this number of cycles is probably too low to achieve a realistic ageing effect (Gale and Darvell, 1999). Recent studies in orthodontics have used various number of thermocycles: approximately 1500 cycles between 10 and 50°C after 3 months of storage (Trites *et al.*, 2004), 500 cycles between 5 and 55°C (Bishara *et al.*, 2007), and 6000 cycles between 5 and 55°C (Faltermeier *et al.*, 2007).

The purpose of this study was to investigate the effects of long-term storage (2 years) and thermocycling (6000 iterations) on the SBS of two SEP adhesive systems. The null hypothesis tested was that the SBS of the self-etching adhesive systems would decrease with long-term storage and thermocycling.

Materials and Methods

One hundred and twenty-six non-carious human maxillary premolars were used in this study. The teeth, which had been extracted for orthodontic reasons, were randomly divided into nine groups of 14 specimens for measurement of SBS. Selection criteria included the absence of any visible decalcification or cracking of the enamel surface under a stereomicroscope (SEM; SMZ 1500, Nikon, Tokyo, Japan) at a magnification of ×10. The extracted teeth were stored in a 0.5 per cent chloramine solution at approximately 4°C. The buccal surfaces of all teeth were cleaned using non-fluoridated pumice. The teeth were also polished using a rubber cup, thoroughly washed, and dried using a moisture-free air source.

Group 1: Transbond XT etch and rinse adhesive system. The enamel surfaces were treated with 35 per cent phosphoric acid etching gel (Transbond XT Etching Gel, 3M Unitek) for 15 seconds, washed for 20 seconds, and dried with oil-free air stream. Table 1 lists the bonding materials used in the present study and Figure 1 is a flow chart of the bracket-bonding instructions. Transbond XT primer was applied to the etched surface, and metal upper premolar brackets (Victory Series, 3M Unitek), with a base area of 10.0 mm^2, were bonded with Transbond XT composite (3M Unitek).

Group 2: Transbond Plus SEP adhesive system. Transbond Plus SEP (3M Unitek) was applied and rubbed on the enamel surfaces for approximately 3 seconds. An air jet was lightly applied to the enamel, and the brackets were bonded with Transbond XT composite.

Group 3: Beauty Ortho Bond SEP adhesive system. Beauty Ortho Bond primers A and B (Shofu) were mixed. The solution was then rubbed onto the enamel surfaces for approximately 3 seconds. An air jet was briefly applied to the enamel, and the brackets were bonded with Beauty Ortho Bond Paste (composite).

Each bonding procedure was performed by the same operator (MI). The excess bonding material was removed with a small scaler. All samples were light cured for 20 seconds (Jetlite 3000, J.Morita USA Inc., Irvine, California, USA) (10 seconds from each proximal side).

After bonding, the specimens were stored in artificial saliva at 37°C for 24 hours (T1) or 2 years (T2). A third group (T3) was thermocycled between 5 and 55°C for 6000 cycles after 24 hours of storage at 37°C in distilled water. SBS was then measured. The specimens were fixed to a custom-fabricated acrylic resin block using Model Repair II (Densply-Sankin, Tokyo, Japan) and the block was fixed to a universal testing machine (EZ Test, Shimadzu, Kyoto, Japan). A knife-edged shearing blade was secured to the crosshead with the direction of force parallel to the buccal surface and the bracket base. Force was applied directly to the bracket–tooth interface. The brackets were debonded at a crosshead speed of 0.5 mm/minute.

After bond failure, the bracket bases and enamel surfaces were examined with a SEM at a magnification of ×10. The adhesive remnant index (ARI) scores were used to assess the amount of adhesive left on the enamel surface (Årtun and Bergland, 1984).

The interface morphology between the adhesive resin and the intact enamel was evaluated under a SEM (SSX-550, Shimadzu). After the SBS was determined, the specimens were cut with a slow-speed water-cooled diamond saw (Isomet, Buehler, Lake Bluff, Illinois, USA), so that they were divided into occlusal and cervical halves; one half was encapsulated for observation of the adhesive interface. The specimens were then polished using a series of abrasives, finishing with a 1 μm diamond paste to obtain a suitable polished surface. The encapsulated specimens were immersed in 6 M hydrochloric acid for 40 seconds and then dehydrated in a graded series of ethanol and water up to 100 per cent ethanol. All specimens were sputter coated with gold (SC-701AT, Sanyu Electron, Tokyo, Japan) and examined under a SEM operating at 15 kV.

Table 1 Materials and instruction employed in present study.

Material	Manufacturer	Components (lot no.)	Composition	pH[a]	Instructions
Transbond XT	3M Unitek, Monrovia, California, USA	Etching gel: (6GN); primer: (5CL); paste: (6TG)	35% phosphoric acid, tetraethylene-glycol dimethacrylate (TEGDMA), bisphenol-A-diglycidel methacrylate (Bis-GMA); Bis-GMA, TEGDMA, silane-treated quartz, amorphous silica, camphorquinone	1.39	Etch enamel 15 seconds; rinse and air-dry; apply thin coat primer; apply adhesive to bracket; 20 seconds light curing
Transbond Plus self-etching system	3M Unitek	Self-etching primer: (237956E); paste: (6TG)	Water, methacrylated phosphoric acid, esters, amino benzoate, camphorquinone, Bis-GMA, TEGDMA, silane treated quartz, amorphous silica, camphorquinone	1.85	Apply primer 3 seconds; gentle air-dry; apply adhesive to bracket; 20 seconds light curing
Beauty Ortho Bond self-etching system	Shofu, Kyoto, Japan	Primer A: (030602); primer B: (030602); paste: (120503)	Water, acetone, others, phosphoric acid monomer, ethanol, TEGDMA, surface pre-reacted glass-ionomer, filler, Bis-GMA, camphorquinone	2.20	Apply primer 3 seconds; gentle air-dry; apply adhesive to bracket; 20 seconds light curing

[a]Published values.

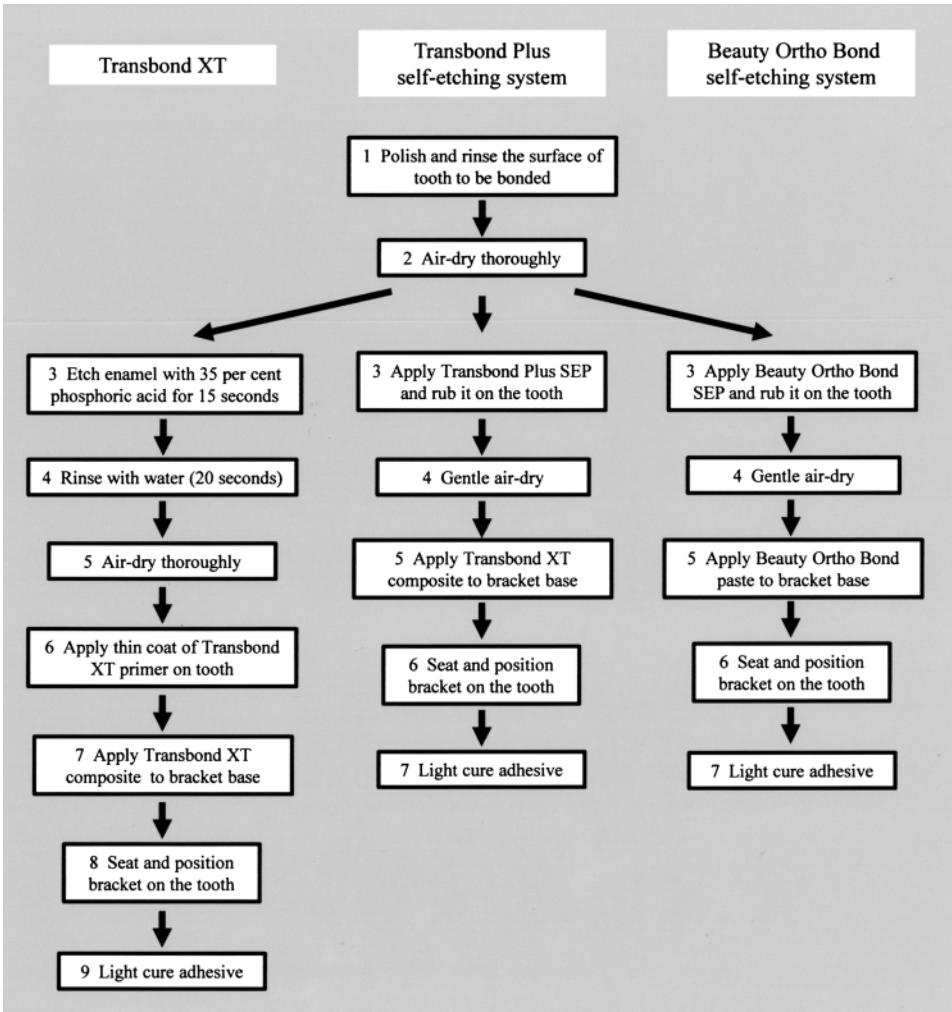

Figure 1 Flow chart of bracket bonding.

Statistical analysis

Statistical analysis was performed using the Statistical Package for Social Sciences (version 14.0J for Windows, SPSS Inc., Chicago, Illinois, USA). The mean SBSs, along with the standard deviation (n = 14), for the groups of bonding materials were compared by two-way analysis of variance (ANOVA). The two factors for ANOVA were bonding material (Transbond XT etch and rinse, Transbond Plus SEP, Beauty Ortho Bond SEP) and storage method (24 hours, 2 years, thermocycling). Chi-square analysis was used to test the significance of differences in the distribution of ARI scores. The level of statistical significance was set at $P < 0.05$.

Results

The SBS results are shown in Figure 2. Two-way ANOVA showed that bonding material (Transbond XT etch and rinse adhesive system, Transbond Plus SEP adhesive system, Beauty Ortho Bond SEP adhesive system; $P = 0.000$) was a statistically significant factor. The storage method (24 hours, 2 years, thermocycling; $P = 0.408$) was not a statistically significant factor. Specimens bonded with Beauty Ortho Bond produced a significantly lower mean SBS (7.4 MPa) than those bonded with Transbond XT etch and rinse (9.8 MPa) or Transbond Plus SEP (9.1 MPa). There was no significant difference in the mean SBS among the three different ageing methods (T1, T2, and T3) for any of the bonding materials.

Chi-square analysis comparing the ARI scores for the three adhesives revealed a significant difference in the distribution of frequencies among the ARI categories for the three adhesive groups at each storage interval (Table 2). Transbond XT etch and rinse and Transbond Plus SEP had a greater frequency of ARI = 1 and 2, except for the Transbond Plus SEP with thermocycling (T3). On the other hand, the Beauty Ortho Bond SEP had a greater frequency of ARI = 2 and 3 for all three storage methods. Both of

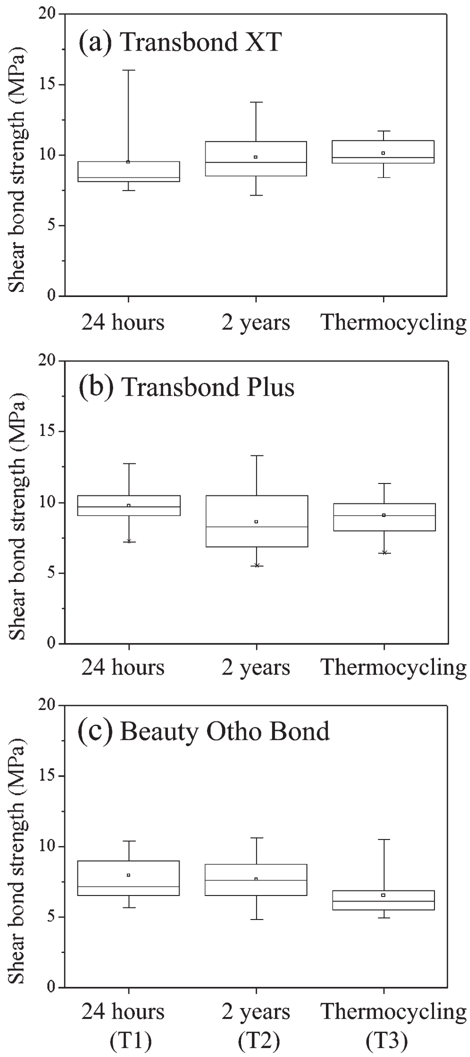

Figure 2 Shear bond strength (MPa) for (a) Transbond XT, (b) Transbond Plus, and (c) Beauty Ortho Bond. Horizontal bars in boxes are medians; 50 per cent of all values are within boxes. The horizontal bars represent the complete range of values. Small boxes within the boxes are average.

ageing methods (T2 and T3) for Transbond XT and Beauty Ortho Bond SEP showed less adhesive remaining on the teeth than the 24 hour storage group (T1).

Figure 3 shows the representative interfaces between the adhesive resin and the intact enamel after SBS testing for T1, T2, and T3, respectively. For Transbond XT, comparatively thick hybrid layers (Nakabayashi and Pashley, 1998), consisting of primer penetrating the surface enamel, were observed with tags ranging from 2 to 10 μm (Figure 3a). On the other hand, a distinct border was seen between the adhesive resin and the enamel for both self-etching adhesive systems (Transbond Plus and Beauty Ortho Bond) and the hybrid layers were less than 2 μm (Figure 3b and 3c). Tight contact between the adhesive resin and the enamel was observed for all specimens after 24 hours of storage (T1) and for the Transbond XT and the Transbond Plus SEP after 2 years of storage (T2) (Figure 3d and 3e) and thermocycling (T3) (Figure 3g and 3h). However, some of the Beauty Ortho Bond specimens showed leakage between the resin adhesive and enamel after 2 years of storage and thermocycling (Figure 3f and 3i).

Table 2 Frequency distribution of adhesive remnant index (ARI) scores.

		ARI scores					Mean
		1	2	3	4	5	
Transbond XT	24 Hours (T1)	10	3	1	—	—	1.4
	2 Years (T2)	4	9	—	1	—	1.9
	Thermocycling (T3)	—	11	3	—	—	2.2
Transbond Plus	24 Hours (T1)	4	6	4	—	—	2.0
	2 Years (T2)	6	6	2	—	—	1.7
	Thermocycling (T3)	1	8	3	1	—	2.4
Beauty Ortho Bond	24 Hours (T1)	4	8	2	—	—	1.9
	2 Years (T2)	1	3	8	2	—	2.8
	Thermocycling (T3)	—	5	6	3	—	2.1

ARI scores: 1, indicates all of the composite, with an impression of the bracket base, remains on the tooth surface; 2, more than 90% of the composite remains on the tooth surface; 3, more than 10% but less than 90% of the composite remains on the tooth surface; 4, less than 10% of composite remains on the tooth surface; 5, no composite remains on the tooth surface.

Discussion

The direct bonding of orthodontic appliances to enamel with acid etching, originally introduced by Newman (1965), has significantly improved the effectiveness of clinical orthodontics. Although acid etching of enamel may remove approximately 10–20 μm of enamel (Shinchi *et al.*, 2000; Powers and Messersmith, 2001), most clinicians accept acid etching of the enamel surface as a routine technique which may cause some iatrogenic effects such as the risk of enamel fracture, surface stains from increased surface porosity, discolouration by resin tags retained in the enamel, and loss of the outer enamel surface (Powers and Messersmith, 2001). Over the past decade, progress has been made in bonding enamel with resin-modified glass ionomers (Cehreli *et al.*, 2005) and SEPs (Cehreli *et al.*, 2005; Bishara *et al.*, 2006), and their lower etching ability might minimize the potential for iatrogenic damage to enamel.

The durability of SEP and resin-modified glass ionomers in clinical use must be evaluated. The most commonly used artificial ageing technique, especially in restorative dentistry, is long-term water storage. Thermocycling is another widely used ageing technique. Bishara *et al.* (2007) studied the effect of ISO standard thermocycling (500 × 5°C/55°C) on SBS for a resin-modified glass ionomer (Fuji Ortho LC) and a SEP adhesive system (Transbond Plus); their mean SBSs after thermocycling were at clinically acceptable levels (6.4 and 6.1 MPa, respectively). However, the number of cycles used in the ISO standard is probably too low to simulate the ageing

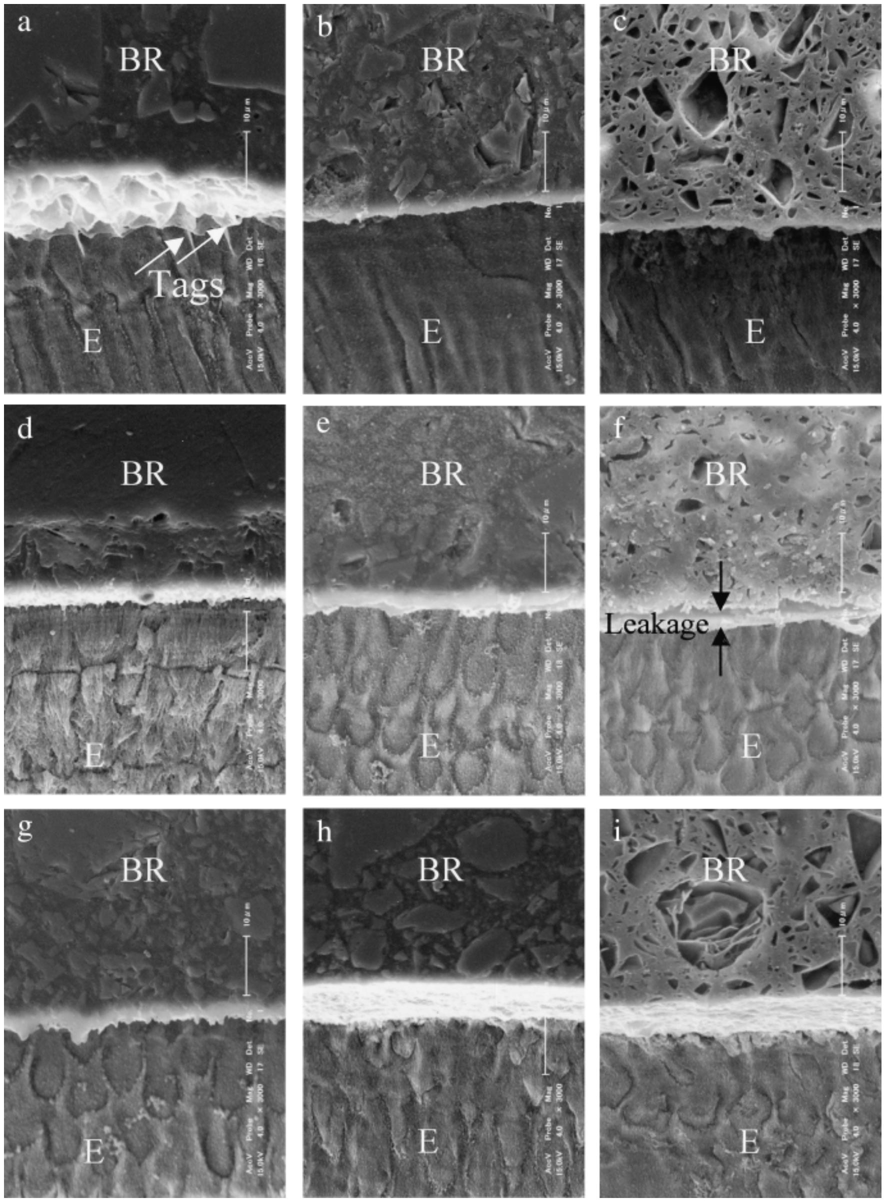

Figure 3 Scanning electron photomicrographs of the adhesive interface between the adhesive resin and the enamel after the shear bond strength (SBS) testing. The specimens were stored in artificial saliva at 37°C for 24 hours (a, Transbond XT; b, Transbond Plus; c, Beauty Ortho Bond), artificial saliva at 37°C for 2 years (d, Transbond XT; e, Transbond Plus; f, Beauty Ortho Bond), or thermocycled between 5 and 55°C for 6000 cycles after 24 hours of storage at 37°C in distilled water (g, Transbond XT; h, Transbond Plus; i, Beauty Ortho Bond) before SBS testing. BR, bonding resin; E, enamel. Magnification ×3000, bar = 10 μm.

effect during long-term orthodontic treatment. Faltermeier *et al.* (2007) compared SBS after thermocycling (6000 × 5°C/55°C) among one-, two- (self-etching systems), and three-component (etch and rinse system) adhesive systems and concluded that there was no significant difference in SBS between the two- and three-component adhesive systems, while the one-component adhesive had a lower bond strength. Most studies have used various types of thermocycling as an artificial ageing technique to understand the durability of bracket bonding but have not used long-term water storage. A decrease in bonding effectiveness is believed to be caused by degradation of the interface components by hydrolysis (Munck *et al.*, 2005). In addition, water can also infiltrate and weaken the mechanical properties of the polymer matrix (Ferracane *et al.*, 1998; Ito *et al.*, 2005). Although it is unclear whether the effect of thermocycling on bond strength is equal to that of long-term storage, limited information is available on the relative effects of thermocycling and long-term storage on bracket bonding. The present study measured SBS after 2 years, which almost corresponds to the duration of orthodontic treatment with fixed appliances and compared the results to bond strength after thermocycling (6000 × 5°C/55°C). There was no significant difference in the mean SBS for any of the bonding materials among the three different storage methods (T1, 24 hours storage; T2, 2 years storage; T3, thermocycling between 5 and 55°C for 6000 cycles). These results confirm that thermocycling is an appropriate method for understanding the durability of orthodontic bracket-bonding materials. All the bonding materials used in the present study with both ageing methods had clinically acceptable levels of SBS.

The ARI scores in the present study showed that Transbond XT and Beauty Ortho Bond SEP adhesive systems had less adhesive remaining on the teeth with T2 and T3 than for the 24 hour storage group (T1). In SEM observation, some of specimens bonded with Beauty Ortho Bond showed leakage between the resin adhesive and enamel after artificial ageing. This may have been due to degradation of the interface components by hydrolysis since mild etching was carried out with a primer with a higher pH. As considerable chair time is needed to remove residual adhesive from the enamel surface, if brackets fail at the enamel–adhesive interface, there would be less residual adhesive, and this might be advantageous if the specimens still had an adequate bond strength.

Conclusions

Under the present study conditions, the following conclusions can be drawn:

1. Thermocycling is an appropriate method for understanding the durability of orthodontic bracket-bonding materials.
2. Both SEP adhesive systems, Transbond Plus and Beauty Ortho Bond, produced adequate SBS even after 2 years of storage and thermocycling between 5 and 55°C for 6000 cycles.

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