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Microtensile Dentin and Enamel Bond Strengths of Recent Self-etching Resins

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Clinical Relevance

Adhesives with self-etching primers, which were evaluated in this study, produced immediate dentin and enamel bond strengths equivalent to the etch-and-rinse product, although they produced less etching of the enamel surface than etching with phosphoric acid.

SUMMARY

In this study, the microtensile bond strengths of resin composites to dentin and enamel produced by recently introduced self-etching resins were determined. Included were two adhesives with self-etching primers, Clearfil SE (Kuraray) and Peak SE (Ultradent), four self-etching adhesives, Optibond All-In-One (Kerr), Clearfil S[3] (Kuraray), Adper Prompt L-Pop (3M ESPE) and iBond (Heraeus Kulzer) and, as a positive control, PQ1 (Ultradent), an etch-and-rinse adhesive. Each product was evaluated using the same hybrid resin composite, Z250 (3M ESPE). Testing was performed after 48 hours using a "non-trimming" microtensile test at a crosshead speed of 0.6 mm/minute. Sample size was five teeth per group, with the value for each tooth calculated by averaging the bond strengths of seven beams derived from it.

Mean values in MPa (SD) for dentin were: Clearfil SE 81.6 (3.5),[a] Peak SE 80.3 (9.9),[a] PQ1 73.4 (4.9),[a,b] Optibond All-In-One 64.4 (5.9),[b] Clearfil S[3] 62.5 (2.2),[b,c] iBond 51.0 (4.0)[c] and Prompt L-Pop 33.9 (6.4).[d] Mean values in MPa (SD) for enamel were: PQ1 55.6 (2.5),[a] Clearfil SE 54.1 (5.4),[a] Prompt L-Pop 54.0 (5.4),[a] Peak SE 51.8 (1.5),[a,b] Clearfil S[3] 44.3 (5.2),[b,c] Optibond All-In-One 40.1 (2.1)[c,d] and iBond 33.8 (3.3).[d] (Values for each substrate with the same letter were not significantly different, one-way ANOVA, Tukey-Kramer Multiple Comparison Test, *p*<0.05.) Compared to

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the positive control, PQ1, only adhesives with self-etching primers, Clearfil SE and Peak SE, were as effective in bonding to both enamel and dentin. With the exception of Prompt L-Pop, scanning electron micrographs of the etched enamel surface produced by self-etching products indicated far less surface topography than conventional etching, even for self-etching primer systems producing the same bond strengths as the etch-and-rinse adhesive.

INTRODUCTION

Current dental adhesives are of three types: etch-and-rinse adhesives, adhesives with a self-etching primer and single-component self-etching adhesives. The latter, although the fastest and most convenient to use, have consistently demonstrated poorer clinical performance in the restoration of unprepared non-carious Class V defects than adhesives with self-etching primers or etch-and-rinse adhesives.[1] This has been attributed to single-component self-etching adhesives being too hydrophilic and susceptible to degradation within dentin hybrid layers[2-4] and to ineffective enamel adhesion, either from inadequate etching of enamel or the poor polymerization of resin once it is infiltrated into the etched surface.[5-6]

Such shortcomings have led manufacturers to retain or introduce adhesive systems that employ self-etching primers, despite their being somewhat less convenient than self-etching adhesives. These shortcomings have also led to the introduction of new self-etching adhesives and to the reformulation of many existing products. Although less compelling than clinical studies, a laboratory screening of initial dentin and enamel bond strengths, which was the purpose of this study, can quickly assess potential improvements in such products.

METHODS AND MATERIALS

Extracted teeth were collected in accordance with human subjects' regulations at the Medical College of Georgia, Augusta, GA, USA. Unerupted third molars stored in 0.9% sodium chloride/0.2% sodium azide at 4°C were used within one month of extraction. For dentin adhesion testing, the occlusal enamel of each tooth was removed using a slow-speed water-cooled saw equipped with a diamond-impregnated disk (Isomet, Buehler, Ltd, Lake Bluff, IL, USA), followed by hand-finishing for 30 seconds on wet 320-grit silicon carbide paper to create a realistic smear layer on the surface of the occlusal mid-coronal dentin. For enamel adhesion testing, flat surfaces were prepared on the facial and lingual surface enamel above the height of contour using 320-grit silicon carbide paper under running water at 60 rpm on a polishing machine (Ecomet 3, Buehler, Ltd).

The adhesives with self-etching primers selected for this study were Clearfil SE Bond (Kuraray Co, Ltd, Osaka, Japan) and Peak SE (Ultradent Products, Inc, South Jordan, UT, USA); the self-etching adhesives were Optibond All-In-One (Kerr Corporation, Orange, CA, USA), Clearfil S^3 (Kuraray Co, Ltd), Adper Prompt L-Pop (3M ESPE, St Paul, MN, USA) and iBond (Heraeus Kulzer, Inc, Armonk, NY, USA) and, as a positive control, PQ1 (Ultradent Products, Inc), an etch-and-rinse adhesive, was included. The adhesives were randomly assigned to 10 extracted teeth, half for enamel and the other half for dentin adhesion testing. Following application of the adhesives according to the manufacturers' instructions, listed in Table 1, 6 mm thick buildups of the hybrid resin composite Filtek Z250 (3M ESPE) were placed. Increment thickness was limited to 2 mm, and curing was accomplished from all directions using a fast halogen light source (VIP, BISCO, Inc, Schaumburg, IL, USA) for a total of five minutes of curing per tooth. Light output was verified to be 600 mW/cm^2 throughout the study, using the unit's built-in radiometer.

After storage in distilled water at 37°C for 24 hours, the restored teeth were sectioned occluso-gingivally into serial slabs approximately 0.8 mm thick using the same slow-speed water-cooled diamond saw. Each slab was then sectioned by the same method into resin composite/tooth structure beams approximately 0.8 x 0.8 mm in cross section, according to the "non-trimming" version of the microtensile test. Each restored tooth yielded 10 to 12 beams for bond strength evaluation, of which seven were chosen at random for testing, for a total of 35 beams for each adhesive/tooth structure combination. After 24 hours' storage in distilled water at 37°C, the dimensions of each beam were determined with a digital caliper accurate to ± 5 µm (Absolute Digimatic Model CD 6" CS, Mitutoyo Corp, Kanagawa, Japan). The beams were then affixed to a Ciucchi device (Kuraray Co, Ltd) using Zapit cyanoacrylate glue (Dental Ventures of America, Corona, CA, USA) and tested to failure under tension in a universal testing machine (Vitrodyne V1000, Chatillon, Largo, FL, USA) at a crosshead speed of 0.6 mm/minute. The type of failure was observed at 2.5x magnification and categorized as adhesive, cohesive in tooth structure or mixed.

In addition to microtensile testing, selected fractured interfaces from each group were evaluated microscopically. The enamel of seven additional teeth was also ground as previously described, and the etching component of each adhesive system was applied as directed, except that the self-etching resins were removed with copious absolute ethanol instead of being light cured. These teeth and the selected fractured beams were then dehydrated in ascending ethanol concentrations and sputter-coated. The fractured beams were examined at 80x magnification, and the etched enamel surfaces examined at 2500x - 20000x magnification in a SEM (Model XL30 FEG, Philips Electronics, Eindhoven, The Netherlands) operated at 5-10kV.

Because multiple beams derived from the same tooth may not be independent samples, bond strengths were calculated for each beam, then the mean of the beams for each tooth was used as its value, yielding a sample size of five per material/substrate subgroup. Data obtained in this manner for the 14 subgroups were statistically analyzed using separate one-way ANOVA and Tukey-Kramer Multiple Comparison Tests for enamel and dentin at a 5% significance level. Since the number of beams was the same for each tooth, this ANOVA approach is equivalent to exact statistical methods that incorporate the intra-class correlation among the beams within each subgroup.[7] At a significance level of 5%, and assuming a coefficient of variation of no more than 10%, this sample size could provide a statistical power of 80%, assuming a 20% difference in mean bond strengths between the highest and lowest subgroups and equal spacing among the subgroups.

RESULTS

Data for each subgroup were found to be normally distributed. For dentin, the self-etching primer adhesives Clearfil SE and Peak SE demonstrated significantly higher microtensile bond strengths than any of the self-etching adhesives and were not significantly different from each other. The bond strength of the etch-and-rinse product PQ1 was not different from the two self-etching primer adhesives, nor was it different

Table 1: *Manufacturer, Instructions/Technique, Composition and Approximate pH for Each Adhesive*

	Technique	**Composition**	**pH**
PQ1 Lot B24MZ Etch Adhesive (Ultradent, South Jordan, UT, USA)	 Apply 15 seconds, rinse Apply 10 seconds, air dry, light cure	 35% phosphoric acid Bis-GMA, 2-hydroxyethyl methacrylate (HEMA), methacrylic acid, ethanol, silica filler	 1
Peak SE Lot PQ SE 34/PQ Thin 07 Primer Bond (Ultradent, South Jordan, UT, USA)	 Apply 20 seconds, air dry Apply, light cure	 Bis-GMA, HEMA, ethanol, methacrylic acid, water Bis-GMA, HEMA, ethanol, methacrylic acid, silica filler	 1.2
Clearfil SE Bond Lot #61739 Primer Bond (Kuraray, Osaka, Japan)	 Apply 20 seconds, air dry Apply, light cure	 10-methacryloyloxy decyl dihydrogenphosphate (MDP), HEMA, water MDP, Bis-GMA, HEMA	 1.9
Clearfil S³ Bond Lot #00001A (Kuraray, Osaka, Japan)	Apply 20 seconds, air dry, light cure	MDP, Bis-GMA, HEMA, water, ethanol	2.7
Adper Prompt L-Pop Lot #254081 (3M ESPE, St Paul, MN, USA)	Mix, apply with agitation 15 seconds, Air dry, reapply, air dry, light cure	di-HEMA phosphate, bisphenol A diglycidyl ether dimethacrylate, water	1
Filtek Z 250 Lot #20060701 (3M ESPE, St Paul, MN, USA)	(Resin Composite)		
iBond Lot #010075 (Heraeus Kulzer, Armonk, NY, USA)	Apply three coats with agitation, 30 seconds, air dry, light cure	4-META, urethane dimethacrylate, glutaraldehyde, acetone, water	1.6
OptiBond All-In-One Lot #45309 (Kerr, Orange, CA, USA)	Apply two coats with agitation, 20 seconds each, air dry, light cure	glycerol phosphate dimethacrylate, mono- and di-functional methacrylate esters, water, acetone, ethanol	2.5

Table 2: *Resin/Dentin Microtensile Bond Strengths—MPa (SD)*

(Z 250 resin composite) Material	n (teeth)	n (beams)	Bond Strengths	Cohesive* (beams)
Clearfil SE	5	35	81.6 (3.5)[a]	8
Peak SE	5	35	80.3 (9.9)[a]	3
PQ1	5	35	73.4 (4.9)[a,b]	4
Optibond All-in-One	5	35	64.4 (5.9)[b]	5
Clearfil S³	5	35	62.5 (2.2)[b,c]	0
iBond	5	35	51.0 (4.0)[c]	0
Prompt L-Pop	5	35	33.9 (6.4)[d]	0

**Failures in dentin; all other failures adhesive.*
Means with same letter not significantly different, one-way ANOVA, Tukey-Kramer Multiple Comparison Test (p<0.05).

Table 3: *Resin/Enamel Microtensile Bond Strengths—MPa (SD)*

(Z 250 resin composite) Material	n (teeth)	n (beams)	Bond Strengths	a	m-e	c-d
PQ1	5	35	55.6 (2.5)[a]	25	1	9
Clearfil SE	5	35	54.1 (5.4)[a]	17	9	9
Prompt L-Pop	5	35	54.0 (5.4)[a]	22	2	11
Peak SE	5	35	51.8 (3.7)[a,b]	25	0	10
Clearfil S³	5	35	44.3 (5.2)[b,c]	30	1	4
Optibond All-in-One	5	35	40.1 (2.1)[c,d]	32	0	3
iBond	5	35	33.8 (3.3)[d]	22	3	9

Failure types (beams): a = adhesive; m-e = mixed/enamel; c-d = cohesive/dentin or dej
Means with same letter not significantly different, one-way ANOVA, Tukey-Kramer Multiple Comparison Test (p<0.05).

from the self-etching adhesives Optibond All-In-One and Clearfil S³. The self-etching adhesives fell into three overlapping groups, with Optibond All-In-One found not to be different from Clearfil S³, while Clearfil S³ was found to not differ from iBond. All materials produced significantly higher dentin bond strengths than Prompt L-Pop.

For enamel, the highest bond strengths were observed for the etch-and-rinse product PQ1, the self-etching primer adhesives Clearfil SE and Peak SE and the self-etching adhesive Prompt L-Pop, which were not significantly different from each other. The remaining products again fell into three overlapping groups, with Clearfil S³ not different from Peak SE or Optibond All-In-One, which was not different from iBond. Complete bond strength results are presented in Tables 2 and 3.

In up to 50% of the enamel adhesion specimens, especially within the highest-

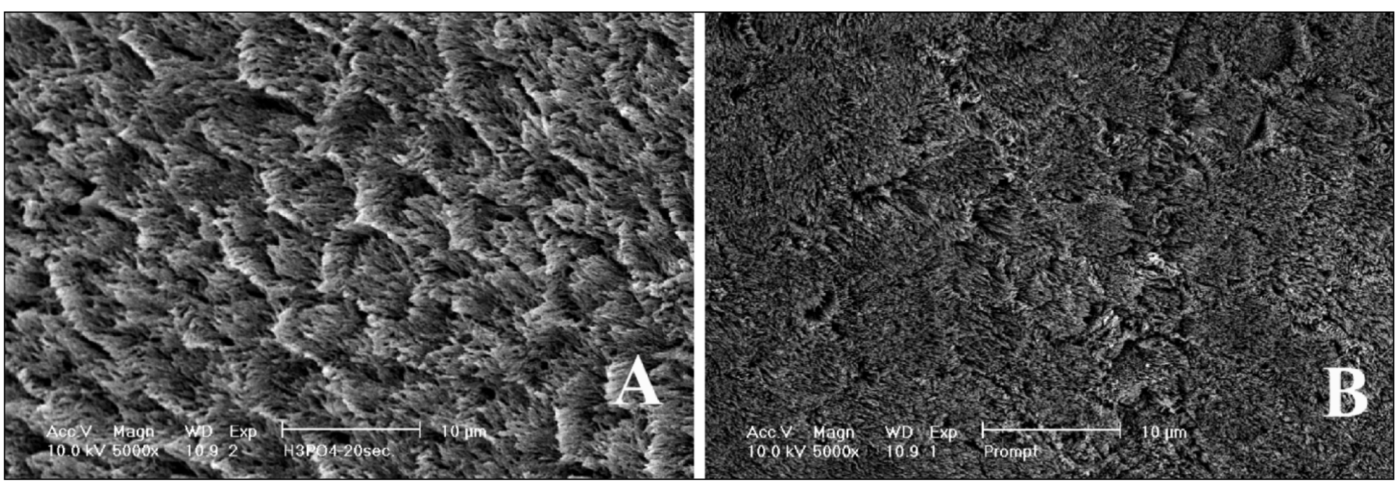

Figure 1. *SEM images of enamel topography produced by (A) conventional phosphoric acid etching, and a relatively acidic self-etching adhesive, Prompt L-Pop (B), which shows nearly equivalent surface etching. (Original magnification = 5000x).*

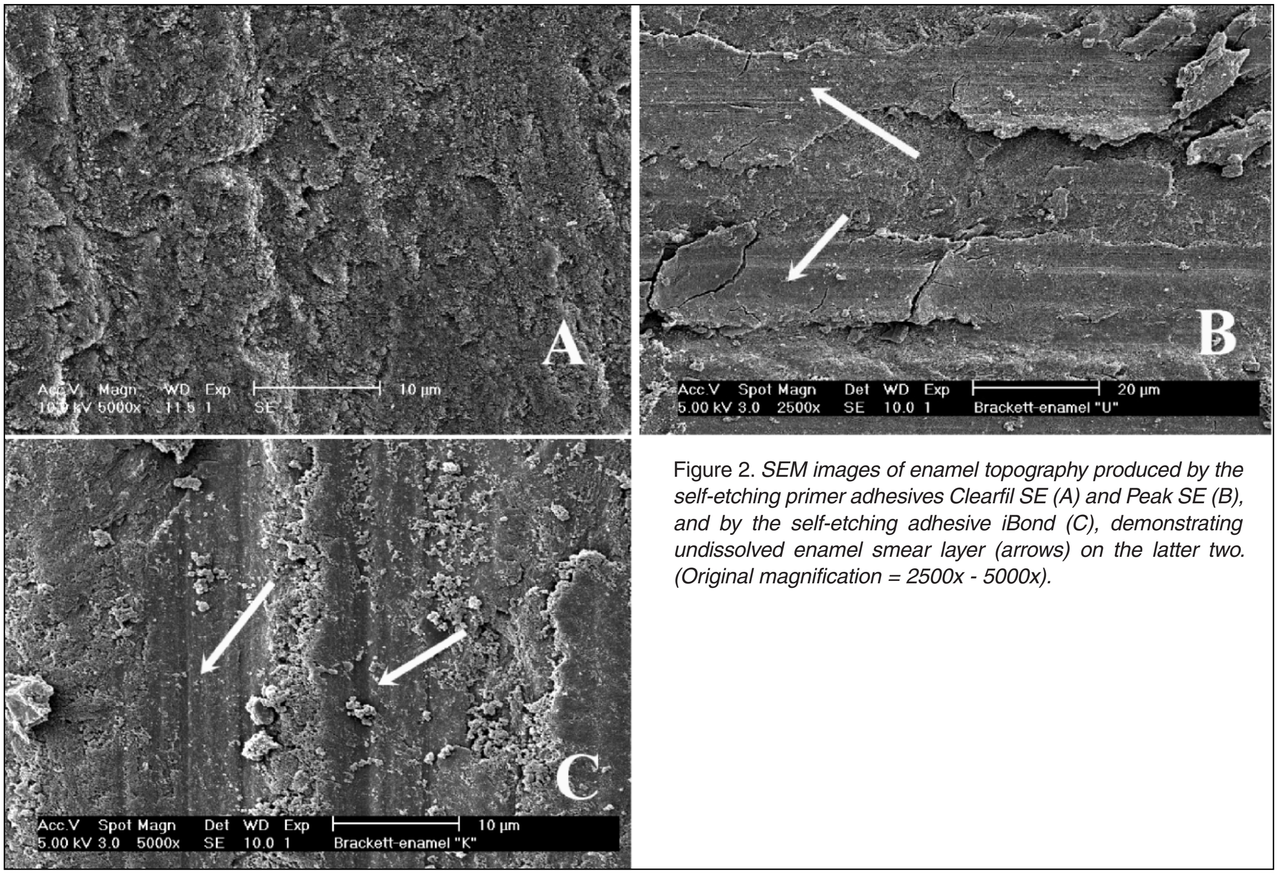

Figure 2. *SEM images of enamel topography produced by the self-etching primer adhesives Clearfil SE (A) and Peak SE (B), and by the self-etching adhesive iBond (C), demonstrating undissolved enamel smear layer (arrows) on the latter two. (Original magnification = 2500x - 5000x).*

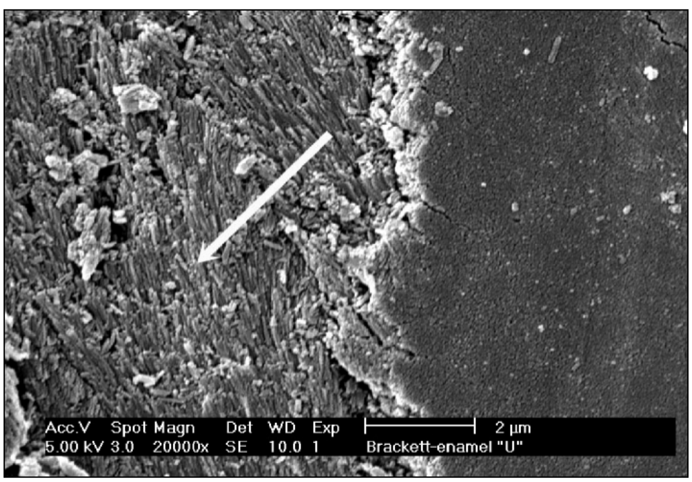

Figure 3. *SEM image at higher magnification of enamel topography produced by the self-etching primer adhesive Peak SE, demonstrating regions of microetched enamel (arrow) between areas of the undissolved enamel smear layer. (Original magnification = 20000x).*

strength groups, failure along the DEJ or within dentin was observed (Table 3). Among the dentin adhesion specimens, up to 25% of the failures occurred within dentin in the highest-strength groups (Table 2).

SEM evaluation showed that conventional phosphoric acid etching and the use of Prompt L-Pop, which has an approximate pH of 1.0, produced the greatest topographical changes in the enamel surface (Figure 1). Among the other self-etching resins, the etched enamel topography was less pronounced, with the undissolved smear layer interspersed with areas of microetched enamel evident for some products, with no apparent correlation found between the surface topography of enamel and bond strength (Figures 2-3). For all the adhesives, there was no obvious correlation between bond strength and microscopic fracture pattern. In general, fractured interfaces appeared to have failed primarily through the unfilled resin layer, with remnants of resin composite attached to tooth structure for most specimens, even those judged visually to have failed adhesively.

DISCUSSION

The most unusual aspect of the current study was that dentin bond strengths were found to be higher than enamel bond strengths, which is the opposite of what is commonly observed in clinical practice. This is most likely attributable to failures of dentin adhesion through hydrolysis occurring in the clinical setting, a process too slow to be discerned by this study, and by the tendency of microtensile enamel bonding specimens to fail along the dentino-enamel junction, which may be weaker than the resin/enamel interface. The latter may cause studies such as this one to understate differences in enamel adhesion, which would be clinically evident. Despite this, the authors believe comparisons of various products within a tissue type, either enamel or dentin, to be valid and discriminating.

Considering specimens from the same tooth to not be independent samples, along with the resultant averaging of the mean for each tooth, produced standard deviations much lower than the approximately 20% of the mean that is usually observed when microtensile specimens are considered independent. Apart from this, little difference would have resulted if the data had been analyzed, with each beam considered an independent specimen, except that less overlapping of the Tukey-Kramer groups would have been evident.

Despite improvements in self-etching adhesives, with some products matching the etch-and-rinse control adhesive in bond strength to either enamel or dentin, only adhesives with self-etching primers compared favorably to the control in adhesion to both substrates. Because cavity preparations and non-carious Class V lesions nearly always include both enamel and dentin interfaces, the authors expect the clinical performance of self-etching adhesives to continue to lag behind that of adhesives that employ a self-etching primer, although not as dramatically as in the past.

It was evident from the SEM images that, even for the adhesives with self-etching primers, much less surface topography was produced on etched enamel surfaces than with conventional phosphoric acid etching. This is probably attributable to the relatively high pH of most self-etching resins relative to 35% phosphoric acid, and it was not evident for the most acidic self-etching adhesive, Prompt L-Pop.

The effectiveness of self-etching primer adhesives in immediate enamel bond strength, despite the lack of surface topography, is probably related to secondary bonding produced through an affinity for calcium, which is evident in some monomers.[8] For Peak SE, which has only recently been introduced, it is not yet known in clinical trials whether this mechanism will be equal to the conventional etching of enamel with phosphoric acid. This has been the case for unprepared Class V restorations retained with Clearfil SE, which have demonstrated 100% retention for up to three years,[9-10] although one of these studies demonstrated less discoloration along the enamel margins over three years when the conventional etching of enamel was employed prior to using Clearfil SE.[9] Concerns over the long-term enamel adhesion of self-etching resins can be addressed through use of this technique, which probably should be used on unground enamel and in instances where enamel adhesion is primarily responsible for retaining restorations that receive significant displacing forces.

Considering the bond strength data and SEM images from this study as a whole, it appears that the milder pH of self-etching primer adhesives is optimal for dentin but may not be sufficiently aggressive for enamel, and there is no single agent/time combination that optimally etches both enamel and dentin.

CONCLUSIONS

Within the limitations of this *in vitro* study, for both enamel and dentin, adhesives with the self-etching primers, Clearfil SE and Peak SE, were as effective in bonding as the positive control, the etch-and-rinse product PQ1. None of the self-etching adhesives was as effective as the control adhesive for both substrates.

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