©Operative Dentistry, 2006, **31-4,** 431-441

Influence of Adhesive Systems on Interfacial Dentin Gap Formation *In Vitro*

AD Loguercio • A Reis • G Bortoli • R Patzlaft
S Kenshima • LE Rodrigues Filho • MLR Accorinte • JWV van Dijken

Clinical Relevance

Etch & rinse systems showed better initial interfacial adaptation than self-etch systems. The differences disappeared after 6-months water storage. The thicker the hybrid layer formed by self-etching adhesives, the lower the immediate gap formation.

SUMMARY

Purpose: This study measured: 1) the interfacial dentin gap formation (IGW) of 2 etch & rinse and 3 self-etch systems with different degrees of acidity and determined the correlation between the smear layer thickness and interfacial gap formation after 1 day and 6 months water storage; 2) the hybrid layer thickness (HLT) produced by the adhesives applied under different smear layer thicknesses. Methods and Materials: Three self-etch adhesives, a mild (Clearfil SE Bond; SE), a moderate (Optibond Solo Plus Self-Etch Primer; SO) and a strong (Tyrian Self Priming Etchant + One Step Plus; TY), and 2 etch & rinse systems (Single Bond; SB and Scotchbond Multi-Purpose Plus; SBMP) were studied. After flat grinding of the occlusal surfaces, 30 molars were sectioned longitudinally in 2 halves. Dentin surfaces with thick and thin smear layers were obtained for each tooth after polishing different sections on 60- and 600-grit SiC paper, respectively. A resin composite (Z250) build-up was made on each tooth section after randomized application of the adhesives. After 24-hour storage in water, resin-dentin bonded sticks (0.8 mm²) were prepared and divided for 1-day and 6-month measurements. The IGW was measured in a stereomicroscope under 400x. SEM micrographs were also made in order to measure the HLT provided by each adhesive. Results: The thickness of the smear layer did not influence the mean gap width (*p*>0.05). The etch & rinse sys-

*Alessandro Dourado Loguercio, DDS, MS, PhD, professor, Universidade do Oeste de Santa Catarina, Campus Joaçaba, Joaçaba, Brazil

Alessandra Reis, DDS, PhD, professor, Universidade do Oeste de Santa Catarina, Campus Joaçaba, Joaçaba, Brazil

Giana Bortoli, undergraduate student, Department of Dental Materials and Operative Dentistry, School of Dentistry, Universidade do Oeste de Santa Catarina, Campus Joaçaba, Joaçaba, Brazil

Rafael Patzlaft, undergraduate student, Department of Dental Materials and Operative Dentistry, School of Dentistry, University of Oeste de Santa Catarina, Joaçaba, SC, Brazil

Silvia Kenshima, DDS, MS, PhD student, Universidade do Oeste de Santa Catarina, Campus Joaçaba, Joaçaba, Brazil

Leonardo Elóy Rodrigues Filho, DDS, MS, PhD, professor, Department of Dental Materials, School of Dentistry, University São Paulo, São Paulo, Brazil

Maria de Lourdes Rodrigues Accorinte, DDS, MS, PhD, clinical practice, São Paulo, Brazil

Jan WV van Dijken, DDS, PhD, professor, Department of Odontology, Dental School Umeå, Umeå University, Sweden

*Reprint request: R Getúlio Vargas, 2125–Bairro Flor da Serra–CEP: 89600-000, Joaçaba/SC, Brazil; e-mail: aloguercio@hotmail.com

DOI: 10.2341/05-53

tems showed the lowest mean IGW in the 1-day group. Their IGW remained unchanged after 6 months. The self-etch systems showed wider initial IGW, which diminished after 6 months water storage, to sizes similar to the etch & rinse systems. The hybrid layer was thicker when bonded to #60 SiC-treated dentin; however, this difference was only statistically different for the 3-step etch & rinse system (*p*=0.001). The thickest hybrid layers were observed for the 2 etch & rinse adhesive systems and the thinnest for the mild self-etch. A negative and strong correlation between IGW and HLT was observed for the self-etching adhesive systems tested (r=80.2, *p*=0.01)

INTRODUCTION

Resin-based bonded esthetic restorative systems are increasingly used in anterior and posterior teeth. Two strategies are currently used to bond resin composites to dentin/enamel: 3- or 2-step etch & rinse systems and 1- or 2-step self-etch systems (Van Meerbeek & others, 2003). The main difference is found in the etching step. In the etch & rinse systems, a separate etching step with phosphoric acid is followed by rinsing with water and careful air drying before application of the primer. Incomplete infiltration of primer monomers into the demineralized dentin is one of the disadvantages of this technique, especially when long etching times are used (Miyazaki, Onose & Moore, 2002; Wang & Spencer, 2003). Incomplete infiltration is partially prevented with the self-etch approach (Carvalho & others, 2005). Demineralization and infiltration occurs simultaneously due to the presence of acidic monomers in the primer.

Controversial results regarding bond strength values of self-etch systems, which may partly be explained by their application on smear layers with varying thicknesses and coarseness (Tay & others, 2000; Tani & Finger, 2002; Oliveira & others, 2003; Kenshima & others, 2005), have been reported. Thick smear layers might affect the ability of self-etch systems to penetrate through intact, mineralized dentin, since early neutralization of the adhesive by dentin buffering components, presented in the smear layer (Oliveira & others, 2003), might hamper superficial demineralization of solid dentin, which is required for collagen exposure. Studies addressing this matter have not reached any conclusion regarding the performance of self-etch systems applied to varied smear layer thicknesses. Some studies reported low resin-dentin bond strengths over thick dentin smear layers (Koibuchi, Yasuda & Nakabayashi, 2001; Miyasaka & Nakabayashi, 1999; Ogata & others, 2001), while others reported no influence of smear layer thickness on resin-dentin bond strengths (Tay & Pashley, 2001; Tani & Finger, 2002; Kenshima & others, 2005).

Bonding systems should reach high initial bond strength to dentin/enamel to be able to counteract the dimensional changes during resin composite polymerization and prevent disruption of marginal sealing. Marginal gap formation will arise if polymerization stresses are higher than the initial bond strength of the adhesive system to tooth tissue (Carvalho & others, 1996; Davidson & Feilzer, 1997). As hybrid layers have a lower elastic modulus than their neighboring substrates, one can theoretically suppose that the thicker the hybrid layer, the lower the initial dentin gap formation, since the hybrid layer will be absorbed by polymerization shrinkage stresses. However, to the extent of the authors' knowledge, no study has addressed the correlation between interfacial gap formation and hybrid layer thickness.

Another important issue regarding gap formation is that hygroscopic expansion of the resin-based materials in the oral environment might reduce or eliminate the size of the marginal gaps caused by polymerization shrinkage of composites (Burrow, Satoh & Tagami, 1996; Thonemann & others, 1997; Huang & others, 2002; Yap, Shaw & Chew, 2003). Water sorption of resin-based materials is related to the characteristics of polymers (Venz & Dickens, 1991), and it is likely that marginal gap reduction also depends on the adhesive system employed.

The resin-bond has been evaluated in the majority of studies by measuring bond strength to dentin or enamel. No correlation has been shown between bond strength values and marginal sealing (Okuda & others, 2001; Guzman-Armstrong, Armstrong & Qian, 2003; Loguercio, Reis & Ballester, 2004; Kenshima & others, 2005). Therefore, a reduction in bond strength does not automatically mean worse marginal adaptation. To date, no study has yet evaluated the sealing ability of self-etch adhesives over time.

The objective of this study was twofold: 1) to determine the initial and 6-month interfacial adaptation of three 2-step self-etch systems with different acidity on dentin with different smear layer thicknesses and 2) evaluate the relationship between hybrid layer thickness and interfacial gap formation. The null hypotheses to be tested were: 1) there is no difference among self-etch and etch & rinse adhesive approaches; 2) interfacial gap formation is not affected by smear layer thickness; 3) there is no difference among self-etch systems with different degrees of acidity; 4) there is no relation between hybrid layer thickness and interfacial gap formation and 5) there is no difference among immediate and 6-month gap width.

METHODS AND MATERIALS

Experimental Design

Forty-five extracted non-carious third molars were used. The teeth were collected after obtaining the patients' informed consent under a protocol approved by the University of São Paulo Institutional Review. The teeth were disinfected in 0.5% chloramine and stored in a saline solution for less than 6 months.

The occlusal surfaces were ground flat to expose dentin using 180-grit SiC paper under water-cooling. The teeth were longitudinally cut in a buccal-lingual direction in 2 sections (halves) (Labcut 1010, Extec Corp, Enfield, CT, USA) (Figure 1). The occlusal surface of one section was then polished on wet 600-grit SiC paper for 60 seconds to obtain a thin smear layer, while the other section was polished with 60-grit SiC to obtain a thick smear layer (Tay & Pashley, 2001). The tooth sections were randomly assigned to 5 adhesive systems. A total of 60 samples were employed, and 6 tooth sections were used in each group.

Three self-etch adhesive systems with different acidities were used: a mild, Clearfil SE Bond (SE, Kuraray Medical Inc, Osaka, Japan; pH≅2); a moderate, Optibond Solo Plus Self-Etch Primer + Optibond Solo Plus (SO, Kerr, Orange, CA, USA; 1<pH<2) and a strong, Tyrian Self Priming Etchant (SPE) + One-Step Plus (TY, BISCO, Schaumburg, IL, USA; pH<1). The pH of the solutions was measured as described by Kenshima and others (2005). Two etch & rinse adhesive systems were used as controls, a 2-step system, Single Bond (SB, 3M ESPE, St Paul, MN, USA) and a 3-step system, Scotchbond Multi Purpose Plus (MP, 3M ESPE). Composition, application mode and batch numbers are described in Table 1.

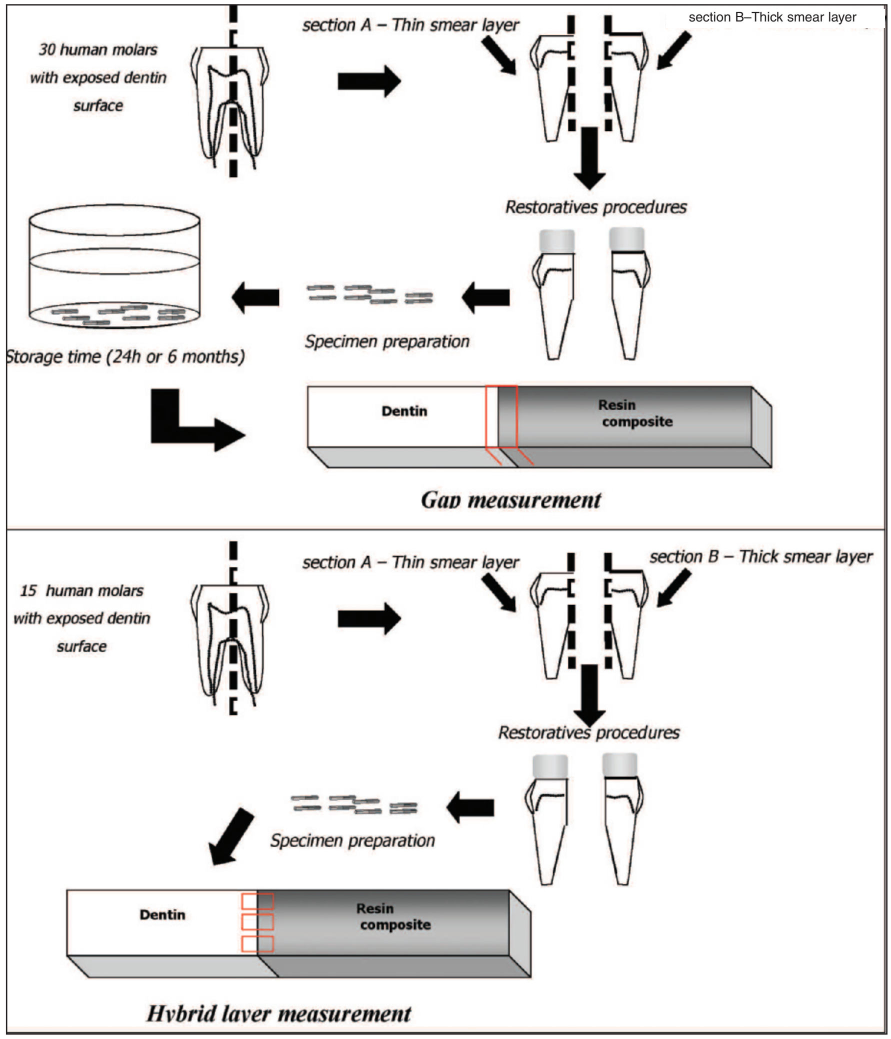

Figure 1. *Schematic representation showing the method to obtain sticks for gap width (above) and hybrid layer thickness (below) measurement. Note the stick in the higher magnification with zones where the hybrid layer thickness was measured in each stick (red square).*

Restorative Procedure

One operator applied all the adhesive systems at 24°C and 50% relative humidity, since these factors may affect adhesive performance (Asmussen & Peutzfeldt, 2003). The adhesives were placed as described in Table 1. Special care was taken to ensure that the dentin surfaces had been adequately covered by primer after evaporation of the solvents. If necessary, additional coats were applied to produce a shiny surface prior to light curing of the adhesives (VIP light-curing unit; BISCO; 600mW/cm^2). A resin composite "crown" (Filtek Z250, 3M ESPE) was prepared with 3 increments approximately 1-mm thick and each was light cured for 30 seconds.

Specimen Preparation and Gap Measurement

After 24-hour storage in distilled water at 37°C, each tooth section was longitudinally sectioned in mesio-to-distal and buccal-to-lingual directions across the bonded interface with a diamond saw (Labcut 1010 machine; Extec Corp, Enfield, CT, USA). Approximately 10 to 12 bonded sticks were obtained for each section, each with a cross-section area approximately 0.8 mm^2. All bonded sticks originating from the same tooth section were randomly divided into 2 parts. One part was evaluated immediately (24 hours), and the second part was evaluated after 6 months of storage in distilled water containing a biocide (0.5% chloramine) at 37°C. The storage solution was not changed and its pH was monitored monthly.

Table 1: *Adhesive Systems and Application Mode*

Adhesive Systems	Composition	Application Mode	Batch #
Clearfil SE Bond—SE (Kuraray)	1. Primer—water, MDP, HEMA, camphoroquinone, hydrophilic dimethacrylate 2. Adhesive—MDP, Bis-GMA, HEMA, camphoroquinone, hydrophobic dimethacrylate, N,N-diethanol p-toluidine bond, silanated colloidal silica	1. Application of 2 coats of the primer with slight agitation (20 seconds); 2. Air-dry (10 seconds at 20 cm); 3. Application of 1 coat of the adhesive (15 seconds); 4. Air-dry (10 seconds at 20 cm); 5. Light-activation (20 seconds–600 mW/cm^2)	00176A 001185A
Optibond Solo Self-Etch Primer + Optibond Solo Plus–SO (Kerr)	1. Alkyl dimethacrylate resins, Barium aluminoborosilicate glass, fumed silica (silicon dioxide), sodium hexafluorosilicate and ethyl alcohol; 2. Alkyl dimethacrylate resins (25-28%), ethyl alcohol, water, stabilizers and activators	1. Application of 1 coat of the primer with slight agitation (15 seconds); 2. Air-dry (10 seconds at a 20 cm); 3. Application of 1 coat of the adhesive (15 seconds with slight agitation); 4. Air-dry (10 seconds at 20 cm); 5. Application of 1 coat of the adhesive (15 seconds with slight agitation); 6. Air-dry (10 seconds at 20 cm); 7. Light-activation (20 seconds–600 mW/cm^2)	205187 203D20
Tyrian SPE and One Step Plus–TY (BISCO)	1. Self-etching primer- 2-Acrylamido-2-methyl propanesulfonic acid (2-15%); Bis-GMA; Ethanol (25-50%) 2. Adhesive– Bis-GMA, BPDM, HEMA, Glass Frit initiator and acetone (40-70%)	1. Mixture of Tyrian SPE (A and B) and application of 2 coats with slight agitation (10 seconds); 2. Air-dry (10 seconds at 20 cm); 3. Application of 2 consecutive coats of the adhesive, brushing for 10 seconds each; 4. Air-dry (10 seconds at 20 cm); 5. Light-activation (10 seconds–600 mW/cm^2)	200002694 200004295
Single Bond–SB (3M ESPE)	1. 37% phosphoric acid 2. Adhesive–Bis-GMA, HEMA, dimethacrylates, polyalknoic acid copolymer, initiators, water and ethanol	1. Acid etching (15 seconds), rinsing (15 seconds) and air-drying (10 seconds), leaving dentin moist; 2. Application of 1 coat of the adhesive (10 seconds with slight agitation); 3. Air-dry (10 seconds at 20 cm); 4. Application of 1 coat of the adhesive (10 seconds with slight agitation); 5. Air-dry (10 seconds at 20 cm); 6. Light-activation (10 seconds–600 mW/cm^2)	2GM
ScotchBond Multi Purpose Plus–SBMP (3M ESPE)	1. 37% phosphoric acid 2. Primer—aqueous solution of HEMA, Polyalkenoic acid copolymer (Vitrebond) 3. Adhesive—Bis-GMA, HEMA, dimethacrylates and initiators	1. Acid etching (15 seconds), rinsing (15 seconds) and air-drying (10 seconds) leaving dentin moist; 2. Application of 2 coats of the primer (10 seconds with slight agitation); 3. Air-dry (10 seconds at 20 cm); 4. Application of 1 coat of the adhesive (10 seconds with slight agitation); 5. Air-dry (10 seconds at 20 cm); 6. Light-activation (10 seconds–600 mW/cm^2)	3008 7543

For both storage times, the gap width at the resin-dentin interface of the bonded sticks was measured with a light microscope (Shimadzu HMV-2, Tokyo, Japan) at 400x magnification (Loguercio & others, 2004). The measurement of the mean gap width of each stick was performed in different rectangular sections with approximately similar adaptation structure. The area of these sections was calculated based on their width and length (Figure 2). The sum of all stick sections, divided by the total length of the interface, resulted in the sticks gap width. The mean gap width of the individual tooth sections was then calculated.

SEM Analysis of the Smear Layer and Hybrid Layer Thickness

In order to verify the thickness of the smear layer produced by the SiC papers, 3 additional teeth for each experimental condition, with dentin exposed by occlusal grinding, were longitudinally sectioned into 2 halves: 1 was polished on 60-grit SiC paper and the other on 600-grit SiC paper. The hemi-sections were mounted on stubs and gold sputtered (MED 010, Balzers Union, Balzers, Liechtenstein) prior to visualization in a scanning electron microscope (LEO 435 VP, LEO Electron Microscopy Ltd, Cambridge, UK). The images were saved, and the smear layer thickness was measured with Software UTHSCSA Image Tool, Version 2.0 (University of Texas Health Science Center, San Antonio, TX, USA).

The thickness of the hybrid layer for each adhesive system was also measured in a similar manner. Three hemi-sections for each experimental condition were used.

The hemi-sections were bonded according to Table 1. Then, each hemi-section was sliced into bonded sticks. The bonded sticks were prepared according to the HMDS drying technique (Perdigão & others, 1995) and embedded in PVC tubes with an epoxy resin (Buehler Ltd, Lake Bluff, IL, USA). After the epoxy resin setting, the thickness of the embedded specimens was approximately half reduced by grinding with silicon carbide papers under running water and by polishing with 1000-grit SiC paper and 6, 3, 1 and 0.25 µm diamond paste (Buehler Ltd). The specimens were ultrasonically cleaned, partially demineralized with HCl 6N (60 seconds) and deproteinized with NaOCl 5% (20 minutes), rinsed, air-dried, mounted on stubs and gold sputtered (MED 010, Balzers Union, Balzers, Liechtenstein). Resin-dentin interfaces, visualized in a scanning electron microscope (LEO 435 VP, LEO Electron Microscopy Ltd, Cambridge, UK), were saved, and the hybrid layer thickness in each stick was measured with the Software UTHSCSA Image Tool. This measurement was performed in 3 regions: left, center and right (Figure 1). The mean hybrid layer thickness of the individual tooth was then calculated.

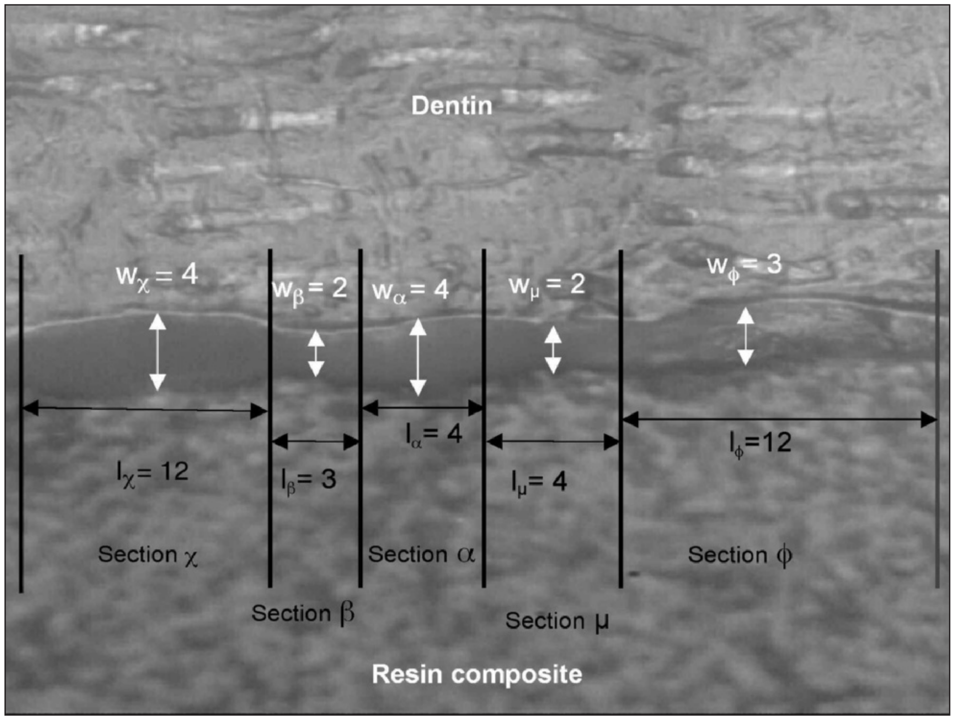

Figure 2. *High magnification of the adhesive interface (400x). For each of the sections (χ, β, α, μ and ϕ), length (l) and width (w) were measured and the area was calculated. For example, the section β area was obtained by multiplication of length l_β by width β w_β: 3 x 2 = 6 µm².*

Statistical Analysis

Statistical Software for Windows (StatSoft, Inc, version 5.0, 1995, Tulsa, OK, USA) was used to process the data. A 3-way repeated measure ANOVA was used to compare the mean gap width (adhesive systems x smear layer thickness x storage time). Time was the repeated factor. An additional random factor was added to the statistical model to correct for multiple samples prepared from the same tooth (DeMunck & others, 2003). Two-way repeated measures ANOVA was employed to analyze the hybrid layer thickness in the experimental groups. Tukey's test was used to compare the mean gap width and hybrid layer thickness between the groups. The level of significance used was p=0.05.

The correlation between the mean gap width of the adhesive systems and their mean hybrid layer thickness were analyzed by simple linear regression analysis. The strength of the association between these two variables was estimated with the Pearson product-moment correlation statistics (α=0.05).

RESULTS

The thickness of the thin and thick smear layer were 1.6 ± 0.6 and 3.2 ± 0.7, respectively. The means and standard deviations of the mean gap width are shown in Tables 2 and 3, respectively. No significant effect of the main factor Smear Layer Thickness was observed on the mean gap (p>0.05). Thus, the summarized data are presented in Table 3, which represents the estimated average mean gap width under a reduced ANOVA model that excluded the factor Smear Layer Thickness. A significant effect of storage time on gap width was found for the Adhesive System (p=0.0001) and interaction Adhesive System X Storage Time (p=0.001). The etch & rinse systems showed the lowest immediate gap width values, which were not statistically changed after 6 months of water storage. The self-etch systems showed statistically wider gaps in the immediate time than the etch & rinse systems. The mean gap width of the self-etch systems was statistically reduced after 6 months of water storage. No significant difference was found among adhesive systems after 6 months storage.

Table 2: *Means, standard deviations (μm) of mean gap width for the experimental groups. The number of sticks tested in each condition is in parenthesis.*

Adhesive Systems	Thickness of the Smear Layer			
	Thin		Thick	
	1-Day	6-Months	1-Day	6-Months
Clearfil SE Bond	5.2 ± 0.9 (25)	1.9 ± 0.5 (17)	3.0 ± 0.5 (22)	1.3 ± 0.4 (15)
Optibond Solo Self-Etch Primer + Optibond Solo Plus	2.5 ± 0.6 (22)	0.8 ± 0.3 (18)	3.2 ± 0.5 (18)	1.1 ± 0.5 (16)
Tyrian SPE + One Step Plus	2.1 ± 0.9 (19)	0.9 ± 0.3 (16)	1.4 ± 0.5 (17)	0.4 ± 0.3 (13)
Scotchbond Multi Purpose Plus	0.8 ± 0.2 (27)	0.8 ± 0.3 (22)	0.9 ± 0.3 (25)	0.6 ± 0.4 (22)
Single Bond	0.6 ± 0.2 (29)	0.7 ± 0.3 (22)	1.0 ± 0.3 (22)	1.0 ± 0.4 (27)

Table 3: *Means, Standards Deviations (μm) and Statistical Significance of Mean Gap Width for the Interaction Adhesives x Time(*)*

Adhesive Systems	Water Storage Time	
	1-Day	6-Months
Clearfil SE Bond	4.1 ± 0.7[a]	1.5 ± 0.5[b,c]
Optibond Solo Self-Etch Primer + Optibond Solo Plus	2.6 ± 0.6[b]	1.0 ± 0.5[c]
Tyrian SPE + One Step Plus	1.6 ± 0.6[b]	0.6 ± 0.2[c]
Scotchbond Multi Purpose Plus	0.8 ± 0.2[c]	0.7 ± 0.2[c]
Single-Bond	0.8 ± 0.5[c]	0.9 ± 0.2[c]

Values with the same superscript letter are statistically similar (p>=0.05).

Table 4: *Means, Standards Deviations (μm) and Statistical Significance of Hybrid Layer Thickness for the Experimental Groups (*)*

Adhesive Systems	Thickness of the Smear Layer	
	Thin	Thick
Clearfil SE Bond	0.5 ± 0.2[d]	1.0 ± 0.2[d]
Optibond Solo Self-Etch Primer + Optibond Solo Plus	0.9 ± 0.2[c,d]	1.2 ± 0.3[c,d]
Tyrian SPE + One Step Plus	1.3 ± 0.4[c]	1.6 ± 0.5[c]
Scotchbond Multi Purpose Plus	3.6 ± 1.1[b]	5.3 ± 0.7[a]
Single-Bond	3.9 ± 1.0[a,b]	4.4 ± 0.7[a]

Values with the same superscript letter are statistically similar (p>=0.05).

The means and standard deviations of the hybrid layer thickness of all adhesives tested under thin and thick smear layer covered dentin are shown in Table 4. A significant effect of the hybrid layer thickness and adhesive systems, as well as the interaction between the main factors, were observed (p=0.01). For all adhesives, the hybrid layer was thicker when bonded to #60 SiC treated dentin; however, this difference was only statistically different for the 3-step etch & rinse system (p=0.001). The thickest hybrid layers were observed for the 2 etch & rinse adhesive systems and the thinnest for the mild self-etch system. The moderate and strong acidic self-etch systems showed similar hybrid layer thicknesses (p>0.05), which were intermediate between the etch & rinse and mild self-etch systems. Representative figures of hybrid layer thickness for each experimental condition are shown in Figures 3 through 7.

The correlation between the mean gap width and hybrid layer thickness, including all adhesives, was significant (p=0.001); however, the Pearson correlation coefficient was low (r=58.6). Then, the correlation was repeated only for the self-etch systems, and a significant negative correlation between mean gap width and hybrid layer thickness (r=80.2, p=0.01) was observed (Figure 8).

DISCUSSION

A major problem with light-cured resin composites is their polymerization shrinkage (Carvalho & others, 1996). The development of polymerization stresses is correlated to the C-factor of the cavity (Feilzer, de Gee & Davidson, 1987). The stresses may cause debonding on the tooth-restorative interface. This situation is even worse in cavities with a non-favorable C-factor, where the gap size is significantly wider than that observed in this investigation (Loguercio & others, 2004). Although the mean gap width for etch & rinse systems was significantly lower than the other systems (leading the authors to reject the first null hypothesis), none of the adhesive systems tested were able to produce entirely gap-free resin-dentin interfaces.

The self-etch adhesives initially showed the widest gaps, irrespective of the dentin smear layer thickness; thus, the second hypothesis was accepted. According to Tay and others (2000), the smear layer is full of easily penetrable subunits with interconnecting channels and is probably not a good barrier for preventing acidic monomers reaching the underlying dentin. Thus,

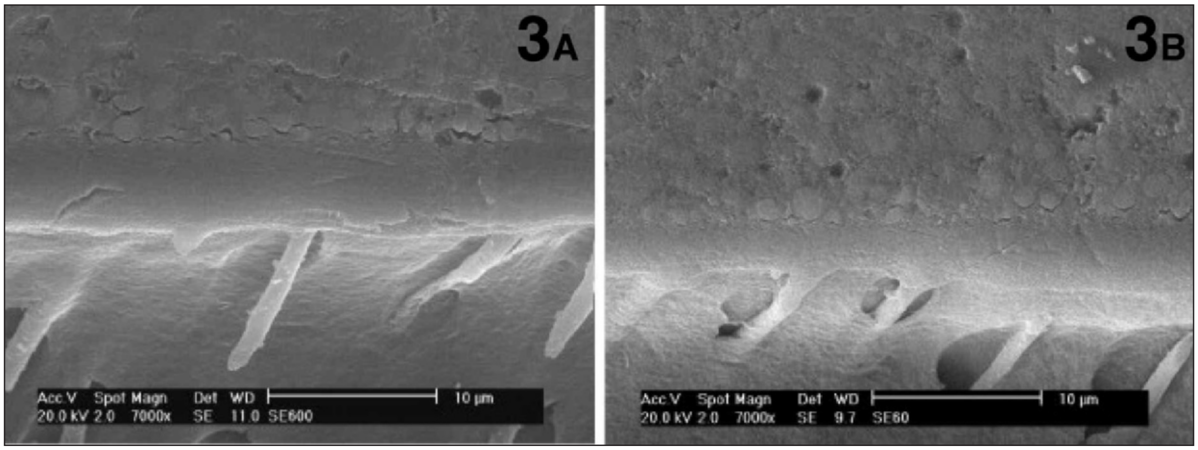

Figure 3. *SEM visualization of resin-dentin interface formed with the mild self-etch system Clearfil SE Bond under thin (3A) and thick (3B) smear layer (7000x magnification).*

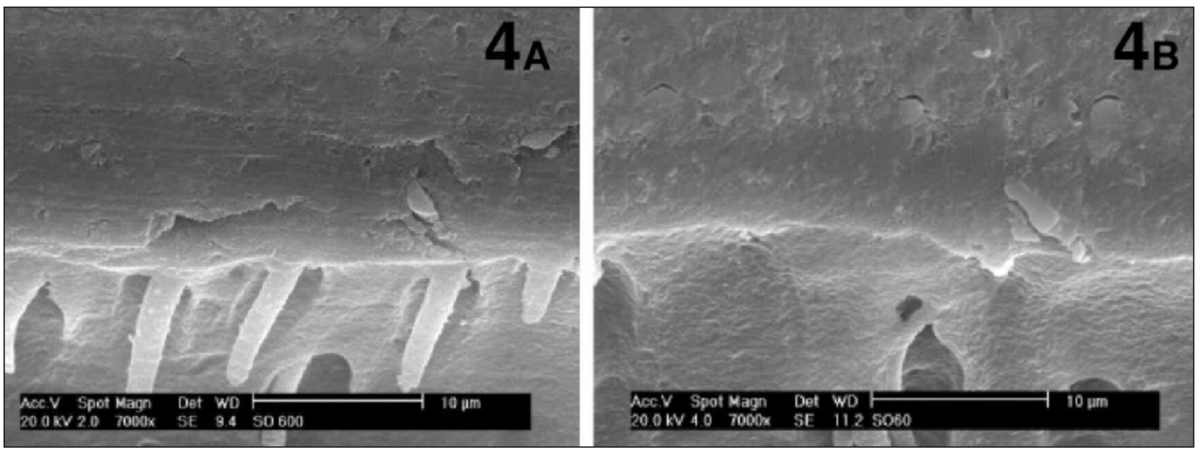

Figure 4. *SEM visualization of resin-dentin interface formed with the moderate self-etch Optibond Solo Self Etch Primer under thin (4A) and thick (4B) smear layer (7000x magnification).*

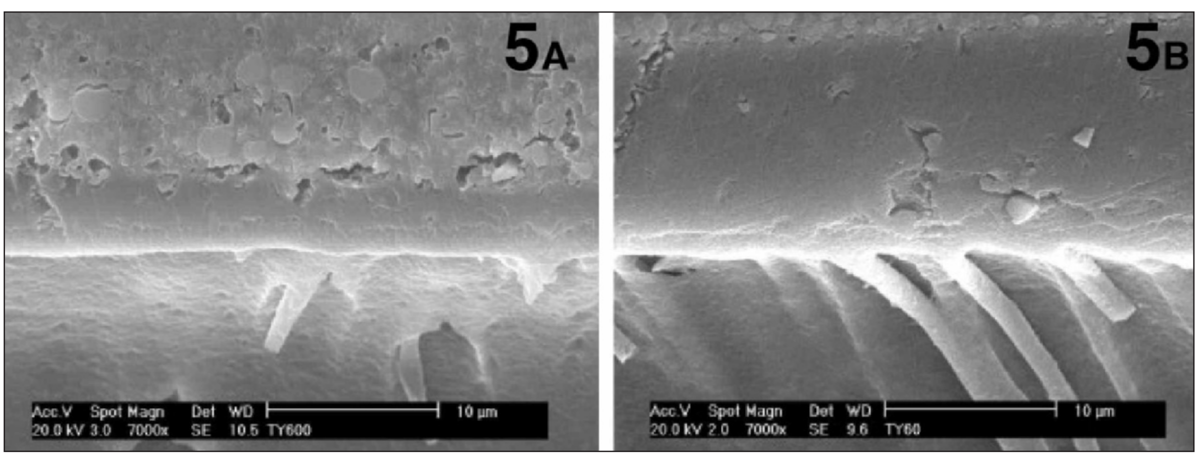

Figure 5. *SEM visualization of resin-dentin interface formed with the strong acidic Tyrian SPE under thin (5A) and thick (5B) smear layer (7000x magnification).*

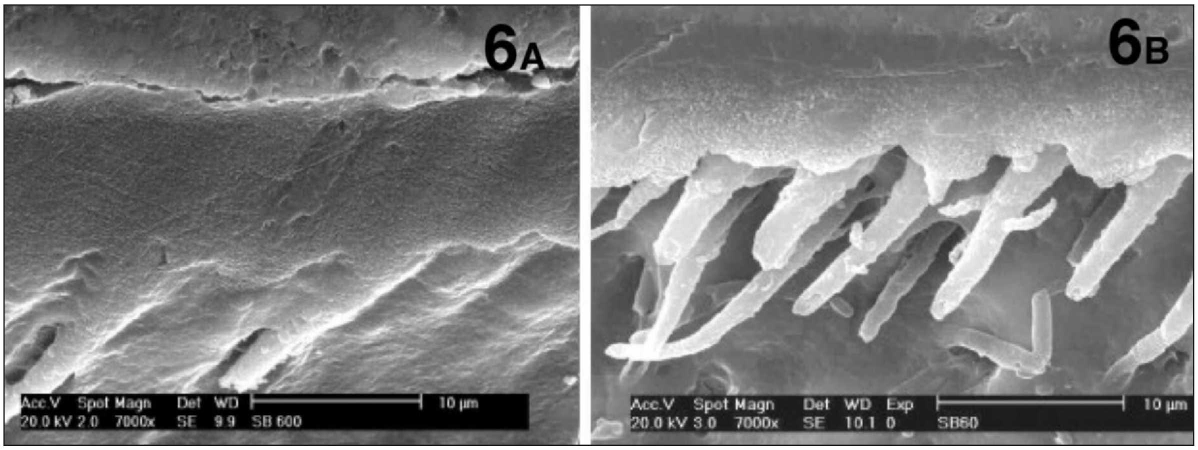

Figure 6. *SEM visualization of resin-dentin interface formed with etch & rinse, 2-step Single Bond under thin (6A) and thick (6B) smear layer (7000x magnification)*

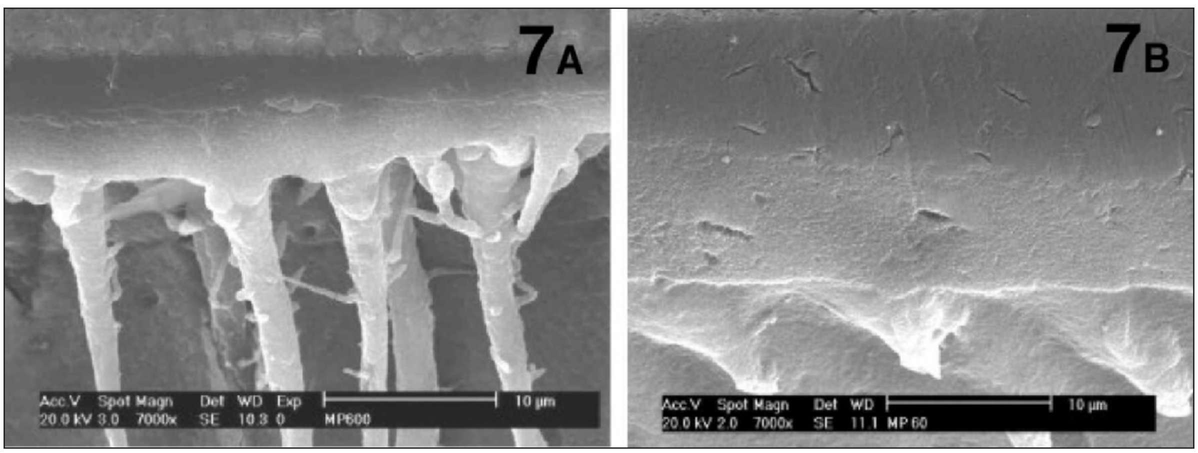

Figure 7. *SEM visualization of resin-dentin interface formed with etch & rinse, 3-step Scotchbond Multi Purpose Plus under thin (7A) and thick (7B) smear layer (7000x magnification).*

acidic monomers can easily penetrate through the channels and promptly reach the underlying dentin. This may explain the lack of significance of the Smear Layer Thickness on gap formation.

The 3 self-etch systems showed significantly different gap widths and the third null hypothesis was rejected. A previous study demonstrated that the depth of penetration of self-etch adhesives into subsurface dentin varies depending on the acidity of the self-etch system (Tay & Pashley, 2001), which is in accordance with this investigation. Thin hybrid layers were formed with the mild self-etch adhesive Clearfil SE Bond; whereas, the moderate and strong self-etch systems formed thicker hybrid layers. However, the hybrid layer thicknesses of the moderate and strong acidic self-etch systems in a study by Tay and Pashley (2001) were 2- to 3-times thicker than the values measured in this study, when applied on thin smear layer dentin. The adhesives employed in this study differ from those evaluated in the study mentioned (Tay & Pashley, 2001); this can solely account for the differences reported. However, it is likely that other factors, apart from adhesive acidity, can be responsible.

The moderate Optibond Solo Self-Etch Primer and the strong self-etch system Tyrian SPE provided lower initial gap width than the mild self-etching Clearfil SE Bond. However, Clearfil SE Bond has shown superior performance in bond strength evaluations compared to more acidic self-etch systems (Kenshima & others, 2005; Sensi & others, 2005). This information, along with previous literature reports, indicates that there is no correlation between bond strengths and hybrid layer thickness (Yoshiyama & others, 1995; Perdigão & others, 2000) and therefore thick hybrid layers could not be considered an additional advantage to obtaining improved retention.

Contrary to the fourth null hypothesis, a negative and significant correlation between the hybrid layer thickness and gap width formation was observed in this study. This suggests that systems with a thicker hybrid layer may provide hybrid layers with improved ability to withstand polymerization stresses and, therefore, restorations with improved sealing. It is known that the hybrid layer possesses an intermediate elastic modulus between mineralized dentin and restorative resin (Van Meerbeek & others, 1993). An increase in hybrid layer thickness does not alter its elastic modulus; however, it increases the resilience of such a layer, that is, this layer can compensate for polymerization stress from the hardening of resin composite. The long-term effect of this finding and its clinical relevance is yet to be addressed.

Both etch & rinse systems showed lower initial gap width compared to self-etch systems. On the other hand, the initial differences in gap width between the 2 adhesive strategies disappeared after 6 months of water storage when all adhesives showed similar gap dimensions, leading the authors to reject the fifth null hypothesis. Resin-based materials are not stable in the wet oral environment. Water storage produces detrimental effects on the mechanical properties of adhesives (Carrilho & others, 2004), with a reduction in the range of 28% to 44% for elastic modulus and ultimate tensile strength. Deterioration of the mechanical properties of some restorative resins and unfilled and filled adhesives has been attributed to the plasticizing effect of water (Beatty & others, 1993). Water sorption causes a softening of the resin component by swelling the polymer network and reducing the frictional forces between polymer chains (Göpferich, 1996; Ferracane & Condon, 1990).

Initially, an outward movement of leachable components, such as solvents and unpolymerized low molecular weight monomers, has been reported, which is followed by continuous water uptake (Mazzaoui & others, 2002; Örtengren & others, 2001a,b; Braden & Davy, 1986). Meanwhile, water uptake reduces the mechanical properties of polymer within the hybrid layer; it also leads to hygroscopic expansion, which compensates partially or totally for interfacial gap formation (Davidson & Feilzer, 1997; Yap & others, 2003).

The role of resin composite material on gap closure has been reported, and its hydrophilic character determines the amount of water uptake in the matrix (Göpferich, 1996; Momoi & McCabe, 1994; Martin, Jedynakiewicz & Fisher, 2003; Yap & others, 2003). Different hydrophilic monomers have been introduced in adhesive systems in years past, in order to optimize the bond to wet demineralized dentin (Kanca, 1992; Reis & others, 2003), and these monomers play an important role in gap closure (Burrow, Inokoshi & Tagami, 1999). In fact, Tanaka and others (1999) reported linear relationships between the amount of water sorption and hydrophilic functional groups of monomers and the reduction of mechanical properties. The self-etch adhesives evaluated in this study showed a high reduction of gap dimension (approximately 60%) after six months of water storage, while gap dimensions of the etch & rinse adhesives remained nearly constant. Self-etch systems contain acidic monomers, water, bifunctional dimetacrylates, HEMA and other bifunctional and multifunctional monomers. Their complex composition, in combination with the inclusion of the smear layer into the hybrid layer, may result in regions of non-optimal conversion within the polymer matrix, partly due to the incomplete removal of solvent.

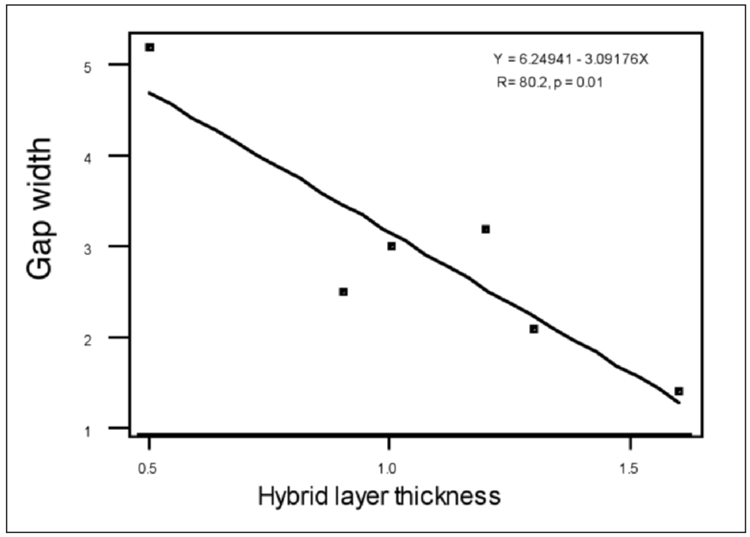

Figure 8. *Correlation and linear relationship between the mean of hybrid layer thickness (μm) and the mean of gap width (μm) for the self-etch adhesive systems tested.*

Tay and others (2002) and Cheong and others (2003) demonstrated the presence of nanopores within the hybrid layer of self-etch adhesives. These nanopores can increase their permeability and allow for the swelling of polymers within the hybrid layer, leading to gap closure. The presence of the hybridized smear layer may be even more important for water diffusion than nanopores, and the hybridized smear layer can also trap more water and solvent within this layer. This would explain why 2-step etch & rinse systems, which also contain nanopores, did not show the same pattern of water sorption as the self-etch systems (Grégoire & others, 2003; Tay & Pashley, 2003). Future studies should be conducted to evaluate these hypotheses.

Water sorption is a slow diffusion-controlled process that continues for several months and compensates partly for the deleterious effects of polymerization shrinkage stresses of resin composite (Sudsangiam &

Van Noort, 1999). It improves marginal adaptation but does not prevent debonding (Davidson & Feilzer, 1997; Huang & others, 2002; Yap & others, 2003). A consequence of water degradation, however, is the reduction of resin-dentin bond strengths after a period of water storage (Hashimoto & others, 2000; Reis & others, 2004).

Water sorption cannot prevent interfacial debonding, which may occur to all adhesives systems even under favorable C-factors, since it cannot counteract the instantaneous shrinkage that occurs on setting (Carvalho & others, 1996; Davidson & Feilzer, 1997). In due course, slight swelling may improve the interfacial adaptation of the restoration but the chances are that, by then, it will be too late. It seems that the first months of service of a composite restoration are the ones in which the restoration is more prone to suffer from microleakage and its consequences. Probably, if appropriate measures are performed in the initial clinical service of a composite restoration, it is likely that resin swelling will improve marginal adaptation in due course.

CONCLUSIONS

It can be concluded that etch & rinse adhesive systems showed better initial interfacial adaptation than self-etch systems. The thickness of the smear layer seems to not affect interfacial gap formation. More acidic self-etch systems resulted in better sealing. Water storage during 6 months improved the interfacial adaptation of self-etch systems to the same quality achieved by etch & rinse adhesives. The thicker the hybrid layer that was formed by the self-etch systems, the lower the immediate interfacial gap formation.

Acknowledgements

The authors are grateful to the Dental School of UNOESC, Joaçaba (SC). This study was partially supported by CAPES and CNPq Grants (551049/2002-2; 350085/2003-0; 302552/2003-0 and 474225-2003-8). The authors thank Dr Carlos Francci for help with the scanning electron microscopy.

(Received 11 April 2005)

References

Asmussen E & Peutzfeldt A (2003) Short- and long-term bonding efficacy of a self-etching, one-step adhesive *Journal of Adhesive Dentistry* **5(1)** 41-45.

Beatty MW, Swartz ML, Moore BK, Phillips RW & Roberts TA (1993) Effect of crosslinking agent content, monomer functionally and repeat unit chemistry on properties of unfilled resins *Journal of Biomedical Materials Research* **27(3)** 403-413.

Braden M & Davy KW (1986) Water absorption characteristics of some unfilled resins *Biomaterials* **7(6)** 474-475.

Burrow MF, Inokoshi S & Tagami J (1999) Water sorption of several bonding resins *American Journal of Dentistry* **12(6)** 295-298.

Burrow MF, Satoh M & Tagami J (1996) Dentin bonding durability after three years using a dentin bonding agent with and without priming *Dental Materials* **12(5)** 302-307.

Carvalho RM, Chersoni S, Frankenberger, R, Pashley DH, Prati C & Tay FR (2005) A challenge to the conventional wisdom that simultaneous etching and resin infiltration always occurs in self-etch adhesives *Biomaterials* **26(9)**1035-1042.

Carvalho RM, Pereira JC, Yoshiyama M & Pashley DH (1996) A review of polymerization contraction: The influence of stress development versus stress relief *Operative Dentistry* **21(1)** 17-24.

Carrilho MRO, Carvalho RM, Tay FR & Pashley DH (2004) Effects of storage media on mechanical properties of adhesive systems *American Journal of Dentistry* **17(2)** 104-108.

Cheong C, King NM, Pashley DH, Ferrari M, Toledano M & Tay FR (2003) Incompatibility of self-etch adhesives with chemical/dual-cured composites: Two-step vs one-step systems *Operative Dentistry* **28(6)** 747-755.

Davidson CL & Feilzer AJ (1997) Polymerization shrinkage and polymerization shrinkage stress in polymer-based restoratives *Journal of Dentistry* **25(6)** 435-440.

DeMunck J, Van Meerbeek B, Yoshida Y, Inoue S, Vargas M, Suzuki K, Lambrechts P & Vanherle G (2003) Four-year water degradation of total-etch adhesives bonded to dentin *Journal of Dental Research* **82(2)** 136-140.

Feilzer AJ, de Gee AJ & Davidson CL (1987) Setting stress in composite resin in relation to configuration of the restoration *Journal of Dental Research* **66(11)** 1636-1639.

Ferracane JL & Condon JR (1990) Rate of elution of leachable components from composite *Dental Materials* **6(4)** 282-287.

Göpferich A (1996) Mechanisms of polymer degradation and erosion *Biomaterials* **17(2)** 103-114.

Grégoire G, Joniot S, Guignes P & Millas A (2003) Dentin permeability: Self-etching and one-bottle dentin bonding systems *Journal of Prosthetic Dentistry* **90(1)** 42-49.

Guzman-Armstrong S, Armstrong SR & Qian F (2003) Relationship between nanoleakage and microtensile bond strength at the resin-dentin interface *Operative Dentistry* **28(1)** 60-66.

Hashimoto M, Ohno H, Kaga M, Endo K, Sano H & Oguchi H (2000) *In vivo* degradation of resin-dentin bonds in humans over 1 to 3 years *Journal of Dental Research* **79(6)** 1385-1391.

Huang C, Tay FR, Cheung GS, Kei LH, Wei SH & Pashley DH (2002) Hygroscopic expansion of a compomer and a composite on artificial gap reduction *Journal of Dentistry* **30(1)** 11-19.

Kanca J 3rd (1992) Effect of resin primer solvents and surface wetness on resin composite bond strength to dentin *American Journal of Dentistry* **5(4)** 213-215.

Kenshima S, Reis A, Uceda-Gómez N, Tancredo de L, Filo LE, Nogueira FN & Loguercio AD (2005) Effect of smear layer thickness and pH of self-etching adhesive systems on the bond strength and gap formation to dentin *Journal of Adhesive Dentistry* **7(2)** 117-126.

Koibuchi H, Yasuda N & Nakabayashi N (2001) Bonding to dentin with a self-etching primer: The effect of smear layers *Dental Materials* **17(2)**122-126.

Loguercio AD, Reis A & Ballester RY (2004) Polymerization shrinkage: Effects of constraint and filling technique in composite restorations *Dental Materials* **20(3)** 236-243.

Martin N, Jedynakiewicz NM & Fisher AC (2003) Hygroscopic expansion and solubility of composite restoratives *Dental Materials* **19(2)** 77-86.

Mazzaoui SA, Burrow MF, Tyas MJ, Rooney FR & Capon RJ (2002) Long-term quantification of the release of monomers from dental resin composites and a resin-modified glass ionomer cement *Journal of Biomedical Materials Research: Applied Biomaterials* **63(3)** 299-205.

Miyasaka K & Nakabayashi N (1999) Combination of EDTA conditioner and phenyl-P/HEMA self-etching primer for bonding to dentin *Dental Materials* **15(3)**153-157.

Miyazaki M, Onose H & Moore BK (2002) Analysis of the dentin-resin interface by use of laser Raman spectroscopy *Dental Materials* **18(8)** 576-580.

Momoi Y & McCabe JF (1994) Hygroscopic expansion of resin based composites during 6 months of water storage *British Dental Journal* **176(3)** 91-96.

Okuda M, Pereira PN, Nakajima M & Tagami J (2001) Relationship between nanoleakage and long-term durability of dentin bonds *Operative Dentistry* **26(5)** 482-490.

Oliveira SS, Pugach MK, Hilton JF, Watanabe LG, Marshall SJ & Marshall GW Jr (2003) The influence of the dentin smear layer on adhesion: A self-etching primer vs a total-etch system *Dental Materials* **19(8)** 758-767.

Ogata M, Harada N, Yamaguchi S, Nakajima M, Pereira PN & Tagami J (2001) Effects of different burs on dentin bond strengths of self-etching primer bonding systems *Operative Dentistry* **26(4)** 375-382.

Örtengren U, Andersson F, Elgh U, Terselius B & Karlsson S (2001a) Influence of pH and storage time on the sorption and solubility behavior of three composite resin materials *Journal of Dentistry* **29(1)** 35-41.

Örtengren U, Wellendorf H, Karlsson S & Ruyter IE (2001b) Water sorption and solubility of dental composites and identification of monomers released in an aqueous environment *Journal of Oral Rehabilitation* **28(12)** 1106-1115.

Perdigão J, May KN Jr, Wilder AD Jr & Lopes M (2000) The effect of depth of dentin demineralization on bond strengths and morphology of the hybrid layer *Operative Dentistry* **25(3)** 186-194.

Perdigão J, Lambrechts P, Van Meerbeek B, Vanherle G & Lopes AL (1995) Field emission SEM comparison of four postfixation drying techniques for human dentin *Journal of Biomedical Materials Research* **29(9)** 1111-1120.

Reis A, Loguercio AD, Azevedo CL, de Carvalho RM, de Julio M & Grande RH (2003) Moisture spectrum of demineralized dentin for adhesive systems with different solvent bases *Journal of Adhesive Dentistry* **5(3)** 183-192.

Reis A, Loguercio AD, Carvalho RM & Grande RH (2004) Durability of resin dentin interfaces: Effects of surface moisture and adhesive solvent component *Dental Materials* **20(7)** 669-676.

Sensi LG, Lopes GC, Monteiro S Jr, Baratieri LN & Vieira LC (2005) Dentin bond strength of self-etching primers/adhesives *Operative Dentistry* **30(1)** 63-68.

Sudsangiam S & van Noort R (1999) Do dentin bond strength tests serve a useful purpose? *Journal of Adhesive Dentistry* **1(1)** 57-67.

Tanaka J, Ishikawa K, Yatani H, Yamashita A & Suzuki K (1999) Correlation of dentin bond durability with water absorption of bonding layer *Dental Materials Journal* **18(1)** 11-18.

Tani C & Finger WJ (2002) Effect of smear layer thickness on bond strength mediated by three all-in-one self-etching priming adhesives *Journal of Adhesive Dentistry* **4(4)** 283-289.

Tay FR & Pashley DH (2001) Aggressiveness of contemporary self-etching systems. I: Depth of penetration beyond dentin smear layers *Dental Materials* **17(4)** 296-308.

Tay FR, Carvalho R Sano H & Pashley DH (2000) Effect of smear layers on the bonding of a self-etching primer to dentin *Journal of Adhesive Dentistry* **2(2)** 99-116.

Tay FR, King NM, Chan KM & Pashley DH (2002) How can nanoleakage occur in self-etching adhesive systems that demineralize and infiltrate simultaneously? *Journal of Adhesive Dentistry* **4(4)** 255-269.

Tay FR & Pashley DH (2003) Have dentin adhesives become too hydrophilic? *Journal of the Canadian Dental Association* **69(11)** 726-731.

Thonemann BM, Federlin M, Schmalz G & Hiller KA (1997) SEM analysis of marginal expansion and gap formation in Class II composite restorations *Dental Materials* **13(3)** 192-197.

Van Meerbeek B, DeMunck J, Yoshida Y, Inoue S, Vargas M, Vijay P, Van Landuyt K, Lambrechts P & Vanherle G (2003) Buonocore Memorial Lecture. Adhesion to enamel and dentin: Current status and future challenges *Operative Dentistry* **28(3)** 215-235.

Van Meerbeek B, Willems G, Celis JP, Roos JR, Braem M, Lambrechts P & Vanherle G (1993) Assessment by nano-indentation of the hardness and elasticity of the resin-dentin bonding area *Journal of Dental Research* **72(10)** 1434-1442.

Venz S & Dickens B (1991) NIR-spectroscopic investigation of water sorption characteristics of dental resins and composites *Journal Biomedical Materials Research* **25(10)** 1231-1248.

Wang Y & Spencer P (2003) Hybridization efficiency of the adhesive/dentin interface with wet bonding *Journal of Dental Research* **82(2)** 141-145.

Yap AU, Shah KC & Chew CL (2003) Marginal gap formation of composites in dentine: Effect of water storage *Journal of Oral Rehabilitation* **30(3)** 236-242.

Yoshiyama M, Carvalho R, Sano H, Horner J, Brewer PD & Pashley DH (1995) Interfacial morphology and strength of bonds made to superficial versus deep dentin *American Journal of Dentistry* **8(6)** 297-302.