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Quantitative Comparison of the Water Permeable Zone Among Four Types of Dental Adhesives Used with a Dual-cured Composite

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Clinical Relevance

When placing a dual-cured composite, variation in the permeability of adhesive interfaces may affect bonding quality and longevity.

SUMMARY

This study compared silver penetration in the adhesive interface among four versions of adhesives from the same manufacturer: OptiBond FL, OptiBond Solo Plus, OptiBond Solo Plus Dual Cure, and OptiBond Solo Plus Self-Etch, when coupled with dual-cured composite, CoreRestore 2 (Kerr). Twenty flat dentin surfaces were prepared using one of the adhesives and bonded with the composite, following the manufacturer's instructions. The surfaces were sectioned into 2-mm slabs and immersed in ammoniacal silver nitrate for 24 hours. Each specimen was exposed to a photodeveloping solution for eight hours and examined with a scanning electron microscope (SEM). The water permeable area occupied by the silver nitrate tracer was determined, and the relative weight of silver was analyzed by wavelength dispersive spectrometry (WDS). The OptiBond FL group had a significantly lower silver content than the other groups ($p<0.0001$). Each group demonstrated different patterns of silver deposition within the adhesive layer and within various features of artifactual fracture from dehydration stress of the SEM. This may be indicative of weak links in the bonded interfaces. Simplified-step adhesives showed increased permeability, which can lead to disruption of coupling with composites.

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DOI: 10.2341/05-40

INTRODUCTION

Recent studies have shown that some simplified-step adhesive systems are not compatible with self- or dual-cured composites, as manifested by their decreased

bond strength (Sanares & others, 2001; O'Keefe & Powers, 2001). One factor that might contribute to this incompatibility is that the adhesives are more acidic in nature, and they tend to deactivate the basic amine catalyst of self- or dual-cured composites. Many simplified-step adhesives are now supplemented with an additional bottle of activator, which contains a ternary catalyst. However, adjunctive use of the activator was reported to be only slightly effective in improving coupling with self- or dual-cured composites (Tay & others, 2003a) or it even decreased bond strength (Say & others, 2005). Another potential factor is increased permeability of the simplified-step adhesives, because increased permeability precludes optimal polymerization and may deteriorate bonding (Cheong & others, 2003).

This study observed water permeable regions within the adhesive joint between dentin and the composite by using a nanoleakage evaluation technique that provides good spatial resolution of submicron defects (Pioch & others, 2001). The water bound spaces were identified with the help of ammoniacal silver nitrate, after allowing its migration within the porous zone, thus inducing an electron microscopic measurable contrast, as demonstrated in studies by Sano and others (1995); Li, Burrow and Tyas (2000). In addition to providing microscopic characterization, silver within the interface was identified by energy dispersive spectrometry (EDS), and a quantifiable comparison was made among the adhesive systems via wavelength dispersive spectrometry (WDS) (Tay, Moulding & Pashley, 1999; Engqvist & others, 2004). This study tested the hypothesis that silver uptake within the bonded interface is greater in simplified-step adhesives than in the conventional three-step adhesive.

METHODS AND MATERIALS

Specimen Preparation

Twenty freshly extracted human molars, collected and stored in 0.10% thymol under IRB protocol 0308-74 (Indiana University and Clarian Institutional Review Boards), were used and flat dentin surfaces were created using a thin sectioning machine (Hamco Machines, Inc, Rochester, NY, USA) (Choeng & others, 2003). The exposed dentin was abraded with wet 240 grit silicon carbide papers (Leco, St Joseph, MI, USA) for 60 seconds to provide clinically relevant thick smear layers suitable for testing self-etch adhesives (Tay & others, 2002). The 20 dentin substrates were randomly divided into four adhesive groups and bonded with one of the adhesives using the manufacturer's instructions included in the packages (Table 1). Both the base and catalyst syringes of CoreRestore 2 were dispensed in equal amounts on the mixing pad and hand mixed for 20 seconds. The composite was applied to form a core 1-mm in height, which was light activated for 20 seconds immediately after placement. After storage in artificial saliva at 37°C for 6 days, each tooth was sectioned occluso-gingivally perpendicular to the adhesive interface along its midpoint with a diamond saw (Isomet 1000, Buehler, Lake Bluff, IL, USA) under water-cooling (Figure 1). From the midline, an additional 2-mm thick section was made, which was coated with nail varnish except for the bonded interface and 1 mm adjacent to the interface (Li Burrow & Tyas, 2001).

Dye Penetration

The specimens were blot dried and immediately immersed in a 50% ammoniacal silver nitrate solution for 24 hours (Pashley & others, 2002). The silver-stained specimens were rinsed in distilled water for 1 minute and placed in a photo-developing solution for 8 hours under a fluorescent light to facilitate reduction of the silver ions into metallic silver particles. All cut surfaces were polished with increasingly fine diamond pastes (0.3 mm, 0.05 mm) and flattened. For observation with SEM, the specimens were cleaned in water for 1 minute, air dried, mounted on aluminum stubs and placed in a desiccator for 24 hours (Li & others, 2000).

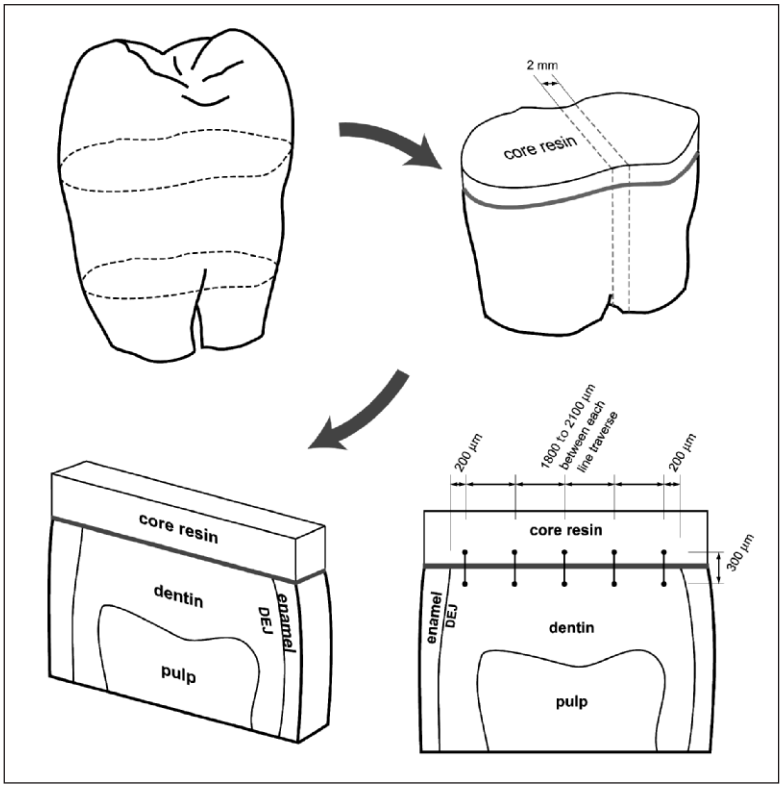

Figure 1. *Schematic illustration of the specimen preparation. The five scan lines were selected perpendicular to the adhesive-composite interface. Each scan line was run from the composite to dentin in a total length of 300 µm, with the same distance to each other.*

SEM Examination with Chemical Analysis

For image and elemental analysis under SEM, the specimens were coated with carbon using a vacuum evaporator (Denton Vacuum DV-502A, Moorestown, NJ, USA). For each specimen, the location where the silver penetrated was identified at 50x to 300x magnification in a Scanning mode of an Electron Probe Micro Analyzer (EPMA; JEOL JXA-8900R, Akishima, Japan) using a phase contrast of back-scattered electron image. The silver weight percents were analyzed with a WDS detector of the same instrument (Tay & others, 1999; Kimura & others, 2001; Hossain & others, 2003). On the image of each specimen, five lines were selected, which were perpendicular to the adhesive-composite interface; the first and last lines were 200 µm away from the dentinoenamel junction. The distance between these lines was identified and divided by four to determine the distance between the middle three lines (Figure 1). Therefore, five scan lines ran from the composite to the dentin, with points of analysis occurring each 0.5 µm (Figure 2). The gap areas produced from the artifactual fractures were regarded as blank spaces and excluded from measurements, because neither silica nor calcium was detected in those areas, both which are representative components comprising the three layers. The values for each 0.5-µm interval were summed and averaged into a single value per scan location. The unit of value was the intensity of the characteristic x-ray per 200 msec, which indicated the relative weight of each element. Operating conditions for both the image and elemental analysis of EPMA were 15kV accelerating voltage, 20nA beam current and 1 mm beam diameter. For elemental analysis, natural apatite and olivine minerals were chosen for the standard of the Ca and Si elements, respectively; whereas, pure silver metal was chosen as the standard for Ag. Repeated measures of analysis of variance (ANOVA) were performed for comparisons between groups at each location and overall, as well as comparisons between locations within each group at a 5% significance level. The analysis had 80% power to detect a 2.8 SD difference between any two groups at any of the five locations.

Table 1: *Adhesives Used in the Study*

Product		Component	Application
OptiBond FL (Kerr, Orange, CA, USA)	Prime	EtOH, CQ,HEMA, GPDM, BHT, water, PAMA	Etch for 15 seconds with 37.5% phosphoric acid (Kerr gel Etchant). Rinse for 15 seconds. Apply Prime and rub for 15 seconds. Dry for 5 seconds. Apply Adhesive in a uniform thin layer. Light cure for 30 seconds.
	Adhesive	Bis-GMA, CQ, HEMA, GDM, ODMAB, S530, A174, OX-50, SP345, Na_2Si_6F	
OptiBond Solo Plus (Kerr)	Prime/ Adhesive	Bis-GMA, HEMA, GDM, GPDM, EtOH, CQ, ODMAB, BHT, TS530,A174, SP345, Na_2Si_6F	Etch for 15 seconds with Kerr gel Etchant. Rinse for 15 seconds. Apply Prime/Adhesive and rub for 15 seconds. Gently air blow for 3 seconds and Light cure for 20 seconds.
OptiBond Solo Plus Dual Cure (Kerr)	Prime/ Adhesive	Same as OptiBond Solo Plus	Mix one drop of OptiBond Solo Plus Prime/Adhesive and one drop of Activator for 3 seconds. Apply the mixture and rub for 15 seconds. Light cure for 20 seconds.
	Activator	Bis-GMA, HEMA, EtOH, BSA	
OptiBond Solo Plus Self-Etch (Kerr)	Self-etch Primer	HFGA-GDM, GPDM, EtOH, MEHQ, ODMAB, CQ	Apply Self-Etch Primer and rub for 15 seconds. Gently air blow for 3 seconds. Apply OptiBond Solo Plus and rub for 15 seconds. Light cure for 20 seconds.

Information given from the Manufacturer (Kerr); A174=gamma-Methacryloxypropyltrimethoxysilane; Bis-GMA=Bis-phenol-A-bis-(2-hydroxy-3-methacryloxypropyl)ether; BHT=2,6-Di-(tert-butyl)-4-methylphenol; BSA= Benzene sulfinic acid sodium salt; CQ = 1,7,7-Trimethylbicyclo-[2,2,1]-hepta-2,3-dione; EtOH=ethanol; GDM = Glycerol dimethacrylate; GPDM= Glycerol Phosphate dimethacrylate; HEMA= 2-Hydroxyethylmethacrylate; HFGA-GDM= Hexafluoroglutaric anhydride-Glyceroldimethacrylate adduct; MEHQ=4-Methoxyphenol; Na_2Si_6F = Disodium hexafluorosilicate; ODMAB = 2-(Ethylhexyl)-4-(dimetylamino)benzoate; OX-50 = Fumed silicon dioxide; PAMA= Phthailic Acid monomethacrylate; SP345= Barium aluminoborosilicate; TS530= Fumed silicon dioxide

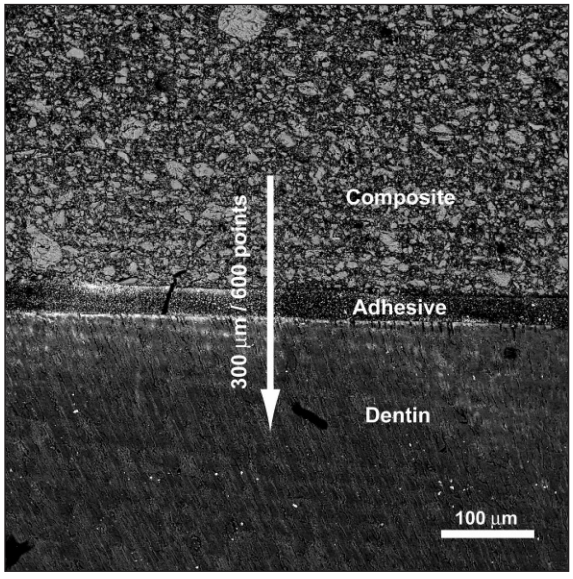

Figure 2. *SEM micrograph (100x) of the adhesive layer of OptiBond FL. The WDS scan line, starting from the composite layer, crosses the adhesive interface and ends in dentin. The scan line has a length of 300 mm, with an interval of 0.5 µm between each point, providing a total of 600 points within the line.*

RESULTS

Each scan line drawn at the five locations for each specimen manifested various silver contents that had been transferred from the dye solution. Line analysis showed

individual distribution of each element, and it was possible to clarify distinct borders in the dentin-adhesive-composite layer (Figures 3a,b,c,d). The adhesive layer begins at the point where the silica contents drop significantly (considering that adhesives are less filled than composite) and ends at the point where the silica falls near zero, and calcium, which is the representative element of dentin, increases. The relative silver contents are shown in Table 2 and Figure 4. Pure silver, used as the standard reference material, has a value of 3766 (Intensity of characteristic x-ray per 200 msec).

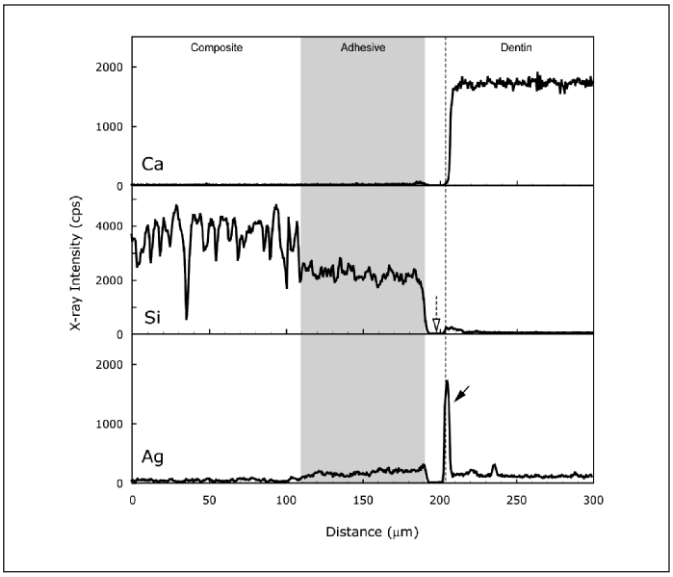

Figure 3a. *Elemental analysis of calcium (Ca), silver (Ag) and silica (Si) on the specimen of the OptiBond FL group. The horizontal scales determine the distance of the scan line across the adhesive interface. The vertical scales are the relative weight value of each material. Artifactual fracture is shown between the adhesive and dentin (broken arrow). There is a slight rise in silver content in the adhesive layer. Note the high peak of Ag as demonstrated in the hybrid layer (arrow).*

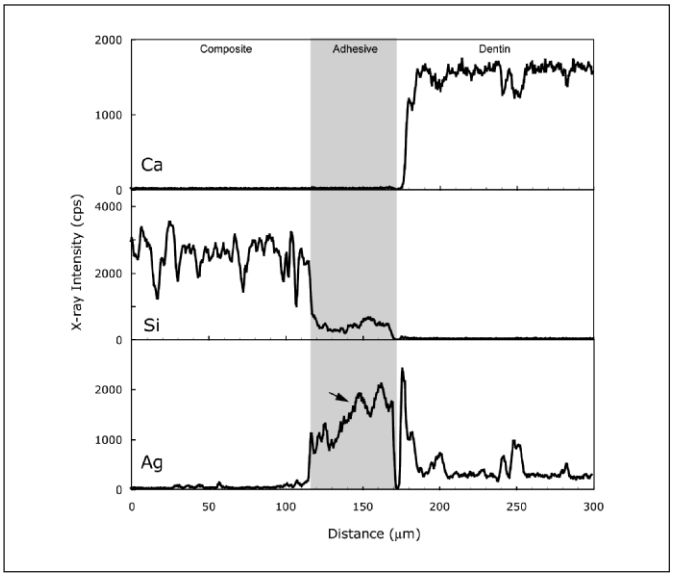

Figure 3b. *Elemental analysis along the specimen of the OptiBond Solo Plus group. Note the silver content rose in the adhesive layer portion (arrow).*

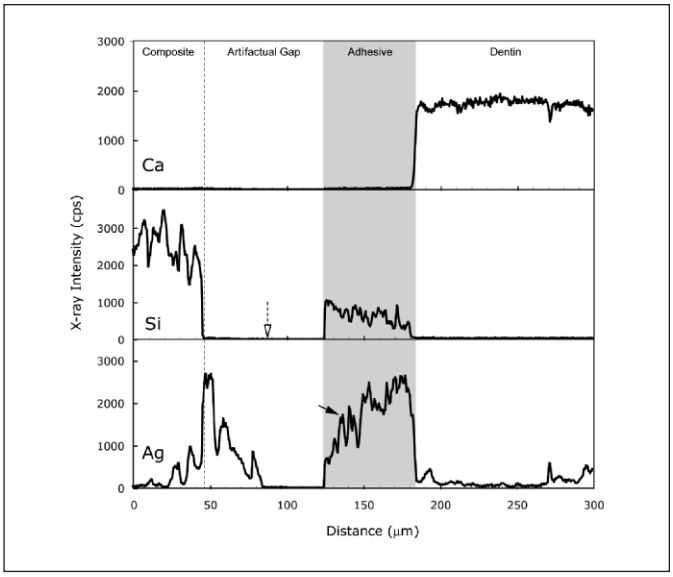

Figure 3c. *Elemental analysis on the specimen of the OptiBond Solo Plus Dual Cure group. In the area where the artifactual gap occurred, silica was not detected (broken arrow). Silver contents were elevated in the adhesive portion (arrow).*

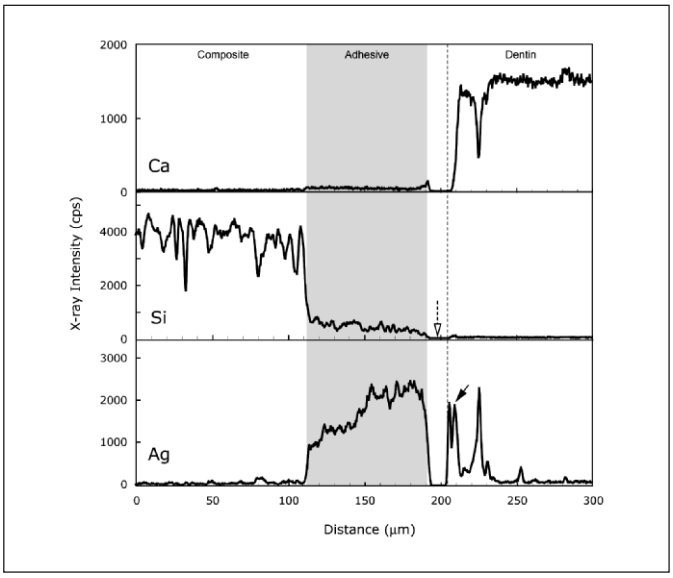

Figure 3d. *Elemental analysis on the specimen of the OptiBond SE group. Note the gap within the adhesive layer from the fracture (broken arrow) near the hybrid layer portion marked with a silver peak (arrow).*

The group-by-location interaction was not significant (p=0.87), indicating comparisons between the locations were similar for each group and comparisons between the groups were similar for each location. No statistically significant differences were found between locations (p=0.22 overall, p=0.82 for OptiBond FL, p=0.99 for OptiBond Solo Plus, p=0.81 for OptiBond Solo Plus Dual Cure, p=0.07 for OptiBond Solo Plus Self-Etch [OptiBond SE, Kerr, Orange, CA, USA]). OptiBond FL had significantly lower silver than OptiBond Solo Plus (p<0.0001), OptiBond Solo Plus Dual Cure (p<0.0001) or OptiBond SE (p<0.0001). OptiBond Solo Plus did not have significantly different silver compared to OptiBond Solo Plus Dual Cure (p=0.51) or OptiBond SE (p=0.94), and OptiBond Solo Plus Dual Cure did not have significantly different silver compared to OptiBond SE (p=0.97).

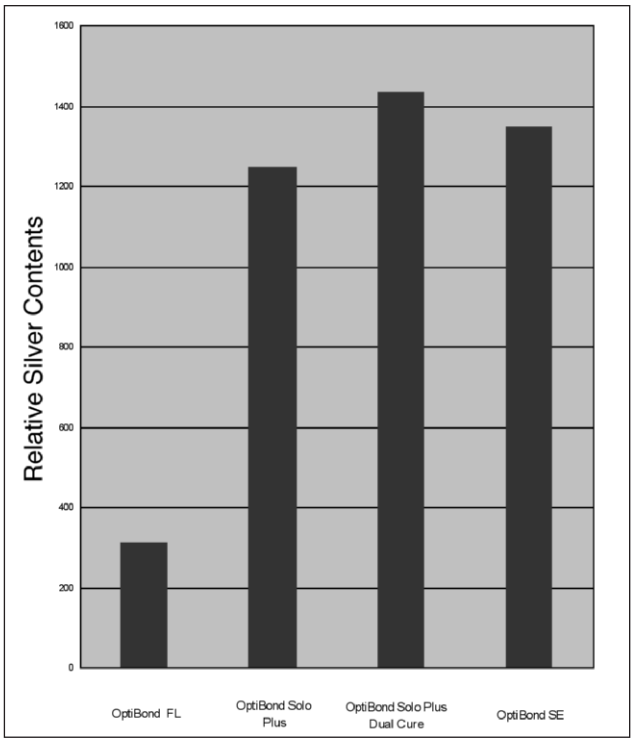

Figure 4. *The mean value of relative silver contents within the adhesive interface of four groups.*

Dehydration stress from the high vacuum of SEM resulted in multiple cracks propagating within the specimens. For the OptiBond FL group, the artifactual cracks occurred in the junction between the adhesive and dentin but did not occur between the adhesive and enamel (Figure 5a). Silver particles were concentrated in the hybrid layer and only presented as a sparse deposition within the adhesive layer, demonstrating a definite margin of the hybrid layer. For OptiBond Solo Plus, silver deposition was more diffuse throughout the adhesive (Figure 5b). Loss of integrity occurred either between the hybrid layer and the adhesive or between the adhesive and the composite (Figure 6a). In OptiBond Solo Plus Dual Cure, crack lines were diverse, with irregular pieces of the fractured adhesive appearing like floating islands (Figure 5c). Fracture lines occurred within the adhesive layer, while the hybrid layer was still intact. For OptiBond SE, the pattern of silver deposition was widespread and fuzzy (Figure 5d). As seen with OptiBond Solo Plus Dual Cure, OptiBond SE also presented with cohesive fractures slightly above the hybrid layer. It is evident that silver particles, discerned by the electro-dense image, coalesced to make droplets, representing the space where water had previously resided (Figure 6b).

DISCUSSION

This study was completed using quantitative measurements that have seldom been accomplished with microscopic observation. Also, the study's protocol followed the manufacturer's instructions and reflected the normal clinical situation, which differs from other *in vitro* studies that have adapting restricted environments (for example, delayed light curing mode, dehydrated dentin) (Tay & others, 2003a; Tay, Pashley & Peters, 2003b; Tay & others, 2004a). From this study, the conventional three-step adhesive OptiBond FL had significantly lower silver contents compared with the other groups. Therefore, the hypothesis that silver uptake within the bonded interface is greater in simplified-step adhesives than in the conventional three-step adhesive, was accepted. OptiBond FL includes a relatively hydrophobic overlying resin coating, unlike the other simplified-step adhesives used in this study. The separate bottle of adhesive in this three-step system provided a more hydrophobic barrier over primed dentin, mini-

Table 2: *Relative Silver Contents Within Adhesive Interface of Four Groups*

Adhesive	Mean	Average Value Per Scan Location				
		Line 1	Line 2	Line 3	Line 4	Line 5
OptiBond FL	309(179)	160(69)[b]	247(231)[b]	369(427)[b]	325(265)[b]	441(220)[b]
OptiBond Solo Plus	1247(173)[a]	1193(443)[c,f]	1318(488)[c,g]	1225(476)[c,h]	1281(500)[c,I]	1218(445)[c,j]
OptiBond Solo Plus Dual Cure	1433(148)[a]	1287(503)[d,f]	1547(132)[d,g]	1440(378)[d,h]	1529(290)[d,I]	1364(228)[d,j]
OptiBond SE	1347(155)[a]	956(508)[e,f]	1292(418)[e,g]	1553(365)[e,h]	1310(476)[e,I]	1624(493)[e,j]

Standard deviations in parentheses. Data with the same letters are not significantly different.

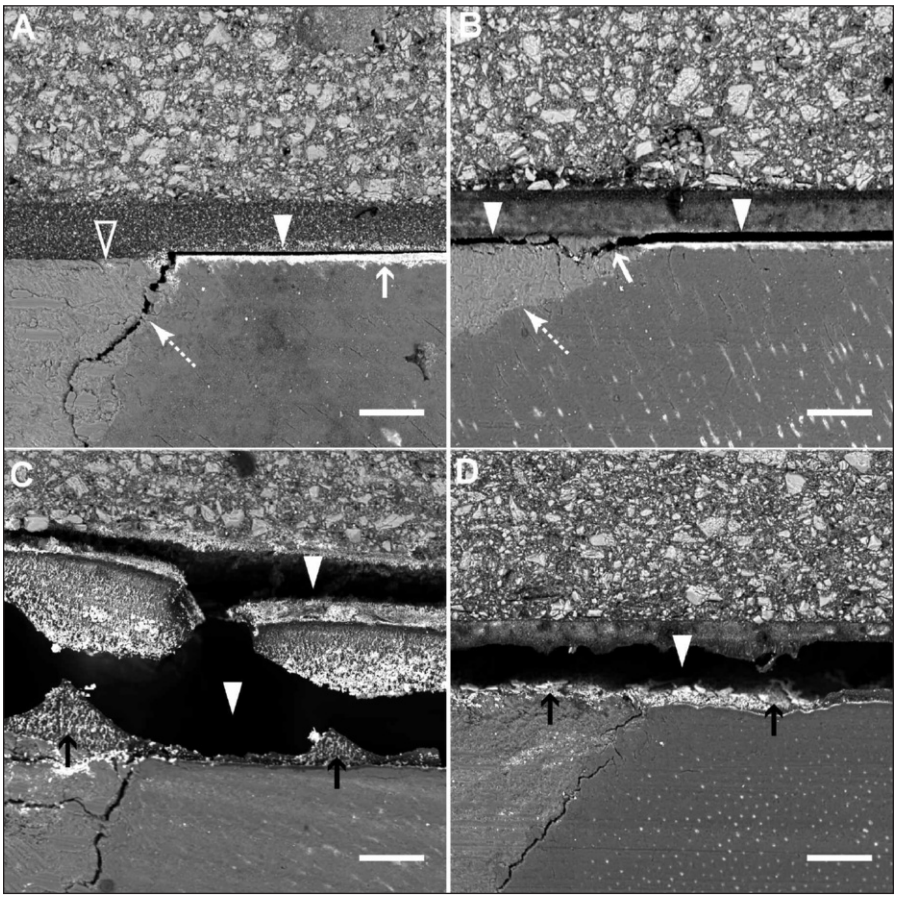

Figure 5. *SEM micrograph (300x) of the adhesive layer. Each scale bar indicates 50 µm. (a) OptiBond FL: the gap between adhesive and dentin (solid triangle) is an artifactual fracture. A fracture line is also evident near the dentinoenomel junction (broken arrow). Silver deposition is mainly localized in the hybrid layer (light arrow). Note the intact enamel bonding (open triangle). (b) OptiBond Solo Plus: artifactual fracture lines (solid triangles) progressed along the adhesive-dentin and adhesive-enamel interface. Enamel bonding is partly retained, giving rise to part of the enamel fracture on that portion (light arrow). The dentinoenamel junction is tightly bound (broken arrow). (c) OptiBond Solo Plus Dual Cure: note the huge space (solid triangles) from the catastrophic fracture within the adhesive and failure of the composite-adhesive bonding. Both the enamel-adhesive and dentin-adhesive interfaces appear intact (arrows). (d) OptiBond SE: an artifactual fracture is seen within the adhesive layer above the hybrid layer (solid triangle). The adhesive interfaces still remain (arrows).*

mizing the chance of water movement. Water, moved from the intrinsically moist dentin structure, is apt to be trapped underneath the adhesive layer that has been light cured immediately after application. This phenomenon was demonstrated by the dense accumulation of silver within the hybrid layer, but only sparsely scattered deposition was found within the adhesive layer (Figure 5a). Conversely, OptiBond Solo Plus, the single-bottle system, and OptiBond SE, the self-etching system, both which introduce the same single bottle of prime/adhesive, showed more silver uptake through the adhesive layer (Figures 5b,d), because they contain hydrophilic regions that allow water to diffuse, while the overlying dual-cure composite is in the process of complete polymerization. These results correlate with recent reports by Tay and Pashley (2003) and Tay and others (2004b) that newly introduced simplified step adhesives are more permeable to water derived from the underlying hydrated dentin, additionally forming multiple water bridges in the intermixed zone between the oxygen-inhibition layer of adhesives and coupling composites during their slow polymerization.

This study introduced EDS for elemental analysis, which requires high vacuum, resulting in artifactual fractures within specimens due to dehydration stress. Interestingly, these unwanted artifacts were likely indicative of where the weak links were in the bonding interfaces. For the OptiBond FL group, fracture lines uniformly propagated along the junction between the adhesive and hybrid layer, because the hybrid layer, manifested by a high peak of silver concentration (Figure 3a), took up water from dentin and acted as a reservoir (Chersoni & others, 2004), eventually playing the role of stress raiser after dehydration. It was remarkable that adhesive-enamel bonding remained intact, dissipating stress into the dentinoenamel junction (Figure 5a). In the OptiBond Solo Plus group, the dentinoenamel junction was mostly preserved, while the bond to enamel was sacrificed (Figure 5b). For dentin bonding, the weak portions varied by location, not only between the adhesive and hybrid, but also between the composite and adhesive (Figure 6a), the latter illustrating osmotic blistering and water droplet accumulation.

OptiBond Solo Plus Dual Cure was supplemented by an additional solution of sulphinic acid sodium salts, which is thought to offset the adverse acid-base reaction to promote its bonding capacity. This activator solution is also comprised of ethanol and 2-hydroxyethly methacrylate (HEMA), which make the adhesive more hydrophilic. In excess water, the monomer phase of HEMA is enriched in polymer and gradually separates into droplets. At the end of the polymerization process, poly[HEMA] becomes filled with large spaces occupied by a water phase (Lou & others, 1999). These water-swollen polymers are supposed to be homogeneous and have inherent porosity. The spaces between the macromolecular chains increase as HEMA polymerizes in the presence of water. By increasing the amount of water in the mix-

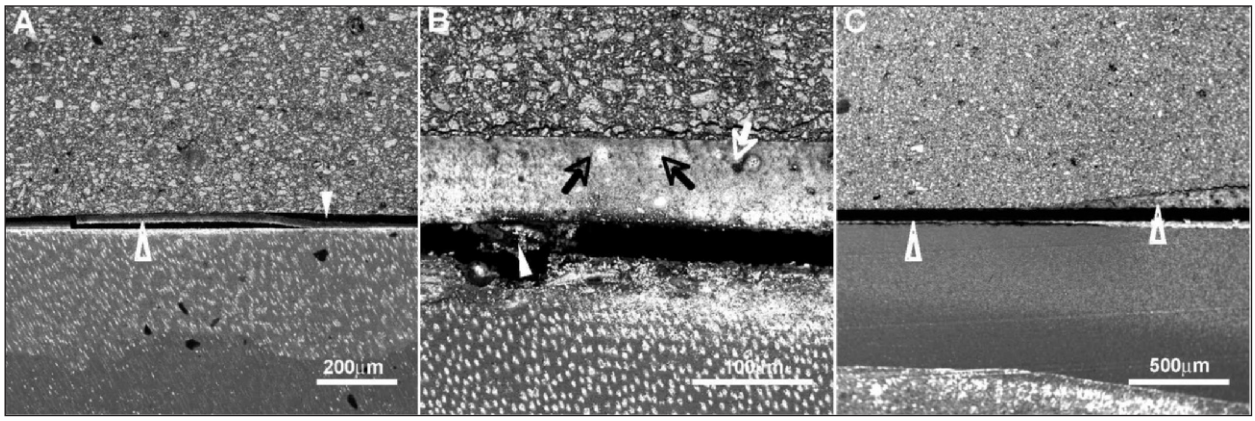

Figure 6. *(a) A low magnification view (100x) of OptiBond Solo Plus showing that an artifactual fracture occurred both on the adhesive-hybrid (open triangle) and the composite-adhesive junction (solid triangle). (b) OptiBond SE containing silver droplets (arrows) at 300x magnification. Interface failure partially occurred in the hybrid layer (solid triangle). The dark circular space may indicate the area with water blisters, but they were not replaced by silver (light arrow). (c) A low magnification view (50x) of OptiBond SE showing the difference in thickness (open triangles). Artifactual fracture occurred within the adhesive interface.*

ture monomer, "microporous" (10 nm to 100 nm) and "macroporous" (100 nm to 1 μm), hydrogels can be produced (Chirila & others, 1993). This extended hydrophilicity and the increased porosity derived from the additional activator might have caused drastic cohesive fracture patterns as well as driven the failure of composite-adhesive bonding as shown in the SEM micrograph (Figure 5c). A similar cohesive fracture pattern was also observed in the OptiBond SE group, but the fracture morphology was relatively uniform, showing that the weak location was right above the adhesive-hybrid interface (Figure 5d). Remarkably, multiple silver depositions in a spherical form appeared throughout the adhesive layer (Figure 6b). It is assumed that the droplet-shaped space had previously been occupied by water and was replaced by silver particles, because univalent ions of silver nitrate compete with water molecules and diffuse rapidly into those spaces (Mair, 1989; Agee & others, 2003; Tay & others, 2004c). The adhesive layer of OptiBond SE varied in thickness at different locations (Figure 6c), because self-etch primer has a very low viscosity and can easily wet the dentin surface. The thickness of OptiBond SE is further affected by the light brushing motion and three-second air thinning used in placement. When a thin consistency of the adhesive is placed over the self-etch primer, the adhesive-primer complex moves laterally, leaving only a microfilm of coating in some areas before it is cured (Özok, 2004).

In currently available adhesives, increased water content after polymerization has been associated with detrimental effects on bonding quality. This study reinforces a previous report (Tay & others, 2003a) that the permeability of simplified-step adhesives contributes to incompatibility with dual-cured composite even though an additional activator governs the adverse acid base reaction. The materials used in the study are produced by the same manufacturer and have been developed in a series of versions for ease of application. However, in coupling with the dual-cured composite, there appeared to be varied water content at microscopic levels.

CONCLUSIONS

Four adhesives from the same manufacturer varied in permeability and showed differing amounts of water content when bonded to a dual-cured composite. Three simplified-systems had more silver tracer uptake within the polymerized adhesive interface than a conventional three-step adhesive.

(Received 20 January 2005)

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