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Adhesive permeability affects coupling of resin cements that utilise self-etching primers to dentine

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Summary *Objectives*. To examine the effects of an experimental bonding technique that reduces the permeability of the adhesive layer on the coupling of resin cements to dentine.

Methods. Extracted human third molars had their mid to deep dentin surface exposed flat by transversally sectioning the crowns. Resin composite overlays were constructed and cemented to the surfaces using either Panavia F (Kuraray) or Bistite II DC (Tokuyama) resin cements mediated by their respective one-step or two-step self-etch adhesives. Experimental groups were prepared in the same way, except that the additional layer of a low-viscosity bonding resin (LVBR, Scotchbond Multi-Purpose Plus, 3M ESPE) was placed on the bonded dentine surface before luting the overlays with the respective resin cements. The bonded assemblies were stored for 24 h in water at 37 °C and subsequently prepared for microtensile bond strength testing. Beams of approximately 0.8 mm^2 were tested in tension at 0.5 mm/min in a universal tester. Fractured surfaces were examined under scanning electron microscopy (SEM). Additional specimens were prepared and examined with TEM using a silver nitrate-staining technique.

Results. Two-way ANOVA showed significant interactions between materials and bonding protocols ($p < 0.05$). When bonded according to manufacturer's directions, Panavia F produced bond strengths that were significantly lower than Bistite II DC ($p < 0.05$). The placement of an additional layer of a LVBR improved significantly the bond strengths of Panavia F ($p < 0.05$), but not of Bistite II DC ($p > 0.05$). SEM observation of the fractured surfaces in Panavia F showed rosette-like features that were exclusive for specimens bonded according to manufacturer's directions. Such features corresponded well with the ultrastructure of the interfaces that showed more nanoleakage associated with the more permeable adhesive interface. The application of the additional layer of the LVBR reduced the amount of silver impregnation for both adhesives suggesting that reduced permeability of the adhesives resulted in improved coupling of the resin cements to dentin.

Conclusions. Placement of an intermediate layer of a LVBR between the bonded dentine surface and the resin cements resulted in improved coupling of Panavia F to dentine.

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Introduction

The advent of adhesive luting cements has considerably expanded the scope of fixed prosthodontics.[1] Bonding of all-ceramic, metal or composite indirect restorations, including fibre posts to root canals are now routine procedures in clinical practice. Clinicians can now select from a wide spectrum of adhesive cements that include water-based materials such as zinc phosphate, zinc polycarboxylate and glass-ionomer cements, the hybrid resin-modified glass-ionomer cements, as well as resin-based compomer and resin composite luting cements.[2,3]

Except for a self-adhesive resin cement (RelyX Unicem, 3M ESPE, St Paul, MN, USA) that does not require pre-treatment of tooth structures, coupling of resin-based cements traditionally requires the adjunctive use of dentin adhesives that are either total-etch or self-etch in nature. The technique sensitivity[4] and the difficulty in achieving a hermetic seal[5] associated with the use of total-etch adhesives probably accounted for the higher incidence of post-operative sensitivity reported with their use in the cementation of indirect restorations.[6] Conversely, resin composite cements that utilise self-etch adhesive components are generally less technique sensitive,[7] and less post-operative cold sensitivity has been reported.[8] It is known that self- or dual-curable resin composites that employ basic amines as part of the redox catalyst are incompatible with the increased concentration of acidic resin monomers utilised in simplified-step dentine adhesives.[9,10] To circumvent this problem, the self-etching primers that are recommended for use with resin cements contain ternary redox initiators such as aryl sulphinate salts, ascorbic acid or barbituric acid salts.[11] This ensures that optimal polymerization of the resin cements occurs when they are used in an auto- or dual-cured mode.

Similar to self-etch adhesives that are marketed for direct restorative procedures, self-etching primers that are manufactured exclusively for use with resin cements may be classified into one-step or two-step systems. Panavia F (Kuraray Medical Inc., Tokyo, Japan) is an example of a one-step self-etch system in which the resin cement is coupled to primed enamel and dentine without an additional resin coating. Bistite II DC (Tokuyama Corp., Tokyo, Japan) is an example of a two-step self-etch system, in which an additional resin coating is placed on top of the primed tooth substrates prior to the application of the dual-cured resin cement. It has been previously reported that one-step self-etch adhesives, because of their higher concentrations of hydrophilic and ionic resin monomers and the lack of the subsequent application of a more hydrophobic resin coating, behave as permeable membranes after polymerization.[12] The increase in permeability in one-step self-etch adhesives allows water to diffuse from dentine across the polymerised adhesive, and form water droplets along the adhesive-composite interface. In the presence of a slow-setting composite, this diffusion process tends to be exacerbated. Increase in adhesive permeability thus provides a second cause of adhesive-composite incompatibility, and is likely to be the major reason for the premature decoupling of dual-cured composites even when ternary redox catalysts are present in the adhesives and/or composites.[10] This may be the reason for the relatively low bond strengths observed when Panavia F was used for luting indirect restorations to hydrated dentine.[13] To overcome this problem, a 'resin coating' technique has been recommended for Panavia F.[14] In this technique, the dentine was sealed with a two-step self-etch adhesive and a light-cured, low viscosity microfilled resin prior to impression taking. Indirect restorations were subsequently luted to this resin-coated tooth surface using Panavia F.

This study examined the effect of adhesive permeability on the coupling of resin cements that employ self-etching primers for bonding to dentine. The rationale behind our study was that if the concept of adhesive permeability is equally applicable to resin cements, the application of an additional coat of more hydrophobic resin to dentine that is treated with the one-step self-etching primer in Panavia F should improve the coupling of this resin cement system to hydrated dentin. Conversely, the use of a similar resin coating would confer less benefit to a resin cement system such as Bistite II DC that already incorporates a two-step self-etching primer. Thus, the null hypothesis tested was that the use of a comparatively more hydrophobic resin coating has no effect on the coupling of both resin cements to hydrated dentine.

Materials and methods

Bonding was performed on non-carious human third molars that were extracted after informed consent had been obtained under a protocol reviewed and approved by the institutional review board from Bauru School of Dentistry USP, Brazil. They were stored in a 1% chloramine T solution at 4 °C and used within one month after extraction. Prior to the bonding experiments, the teeth were retrieved from the disinfectant solution and stored in distilled water, with four changes of the latter within 48 h to remove the disinfectant.

Tooth preparation

Bonding was performed on the occlusal surfaces of mid to deep coronal dentine. The occlusal enamel and the superficial dentine of each tooth were removed using a slow-speed saw (Isomet, Buehler Ltd, Lake Bluff, IL, USA) under water cooling. The tooth surfaces were polished with wet 320-grit silicon carbide abrasive papers to create standard smear layers. The teeth were divided into four experimental groups of seven teeth each. For each group, two teeth were used for transmission electron microscopy (TEM), and the other five teeth for microtensile bond strength (μTBS) evaluation and fractographic analysis using scanning electron microscopy (SEM). The compositions of the two resin cement systems investigated are shown in Table 1. The four experimental groups were:

(a) Panavia F, with dentine treated with the one-step self-etching ED primer, according to the manufacturer's instructions (Table 2).
(b) Panavia F, with the primed dentine covered by a thin layer of a low-viscosity bonding resin (LVBR; Scotchbond Multi-Purpose Plus adhesive, 3M ESPE, St Paul, MN, USA). This adhesive contains Bis-GMA, HEMA and a blend of amine initiators to render it compatible with auto- or dual-cured composites. The resin coating was air thinned and light-cured prior to the coupling of the resin cement.
(c) Bistite II DC, with dentine treated with the two-step self-etching primer, according to the manufacturer's instructions (Table 2).
(d) Bistite II DC, with the primed dentine covered by a layer of LVBR. The bonding resin was similarly air thinned and light-cured prior to the coupling of the resin cement.

Coupling of processed composites

Void-free composite blocks were first produced using a heat- and light-activated hybrid resin composite (Tescera, Bisco Inc., Schaumburg, IL, USA). 5-mm thick layers of composite were dispensed into 2×2 cm^2 flat Teflon moulds (Electron Microscopy Sciences, Fort Washington, PA, USA). The moulds containing the uncured composite were

Table 1 Compositions of the two resin cements investigated in this study.

Resin cement	Components	Composition	Lot number
Panavia F (Kuraray Medical Inc., Tokyo, Japan)		One-step self-etching primer	
	ED primer A	HEMA, MDP, 5-NMSA, water, accelerator	00139B
	ED primer B	5-NMSA, accelerator, water, sodium benzene sulphinate	00025B
		Dual-cured resin cement	
	Universal paste	Hydrophobic aromatic dimethacrylate, hydrophobic aliphatic dimethacrylate, hydrophilic dimethacrylate, sodium aromatic sulfinate (TPBSS), *N*,*N*-diethanol-*p*-toluidine, surface-treated (functionalized) sodium fluoride, silanized barium glass	00102A
	Catalyst paste	MDP, hydrophobic aromatic dimethacrylate, hydrophobic aliphatic dimethacrylate, hydrophilic dimethacrylate, silanized silica, photoinitiator, dibenzoyl peroxide	00035B
		Two-step self-etching primer	
Bistite II DC (Tokuyama Corp., Tokyo, Japan)	Primer 1A	Phosphoric acid monomer, acetone, water	118
	Primer 1B	Initiator, alcohol, water	212
	Primer 2	HEMA, acetone, water	3133
		Dual-cured resin cement	
	Paste A and B	MAC-10, methacrylic monomers, initiators, silica-zirconia filler (77 wt%), Bis-MPEPP, NPGDMA, camphorquinone, initiator	84B-08R

5-NMSA: *N*-methacryloxyl-5-aminosalicyclic acid; Bis-MPEPP: 2,2-bis(4-(methacryloxypolyethoxy)phenyl)propane; HEMA: 2-hydroxyethyl methacylate; MAC-10: methacryloxyundecane dicarboxylic acid; MDP: 10-methacryloyloxydecyl dihydrogen phosphate; NPGDMA: noepentyl glycol dimethacrylate.

Table 2 Application protocol, microtensile bond strengths and failure mode distribution of the two resin cement systems that utilise self-etching primers for coupling of the cement to dentine.

Resin cements	Manufacturer's recommended protocol	Application as per manufacturer's instructions		Light-cured resin coating (Scotchbond Multi-Purpose Plus bonding resin) applied after primer application	
		Bond strength (MPa)[a]	Failure mode	Bond strength (MPa)[a]	Failure mode
Panavia F (1-step)	Apply ED primer to dentine for 60 s, air-dry Apply mixed cement paste, cover with glycerine gel	25.3 ± 8.5^{A} (40)	Mixed failure 40% Adhesive failure 60%	35.6 ± 15.6^{B} (31)	Mixed failure 32.3% Adhesive failure 67.7%
Bistite II DC (2-step)	Apply mixed Primer 1A and 1B for 30 s, air-dry Apply Primer 2 for 20 s, air-dry. Apply mixed cement paste, cover with glycerine gel	33.8 ± 16.0^{B} (47)	Mixed failure 17% Adhesive failure 83%	31.5 ± 12.0^{B} (46)	Mixed failure 54.3% Adhesive failure 45.7%

[a] Values are mean ± standard deviation. Numbers in parentheses represent number of specimen beams employed for microtensile bond testing. Groups with the same letter superscripts are not statistically significant ($P > 0.05$).

placed inside a composite inlay processing chamber (Nitro-Therma-Lite, Bisco Inc.) and light-activated under pressurized nitrogen maintained at 551.6 kPa (i.e. 80 psi) for one complete cycle at 125 °C for 10 min.

After processing, the composite blocks were reduced with the Isomet saw under water cooling to produce smaller blocks that approximate the dimensions of the teeth to be bonded. Each reduced block was then sectioned with the Isomet saw to produce 3 mm thick, parallel-sided composite overlays. The intaglio surface of each composite overlay was sandblasted with 50 μm alumina, cleaned with a phosphoric acid gel (Uni-Etch, Bisco, Inc.), air-dried and silane-treated using RelyX ceramic primer (3M ESPE).

The resin cements were mixed according to respective manufacturer's directions and placed on the treated surface of the composite overlays. The resin blocks were then luted on their respective bonded tooth surface under a 5 kg load that was maintained for 30 s during which the excess cement was carefully removed with a brush. The load was then removed and the dual-cured resin cement (Bistite II DC) was light-cured along the bonded interface (XL3000, 3M ESPE, St Paul, MN, USA, operating at 550 mW/cm^2) at four antagonistic sites for 40 s each. Liquid glycerine gel (Air Barrier for Bistite II DC) was liberally applied around the resin cement margin. For Panavia F, the bonded interfaces were entirely covered with the liquid glycerine gel (Oxyguard II for Panavia F) immediately after the load was removed to enable optimal anaerobic polymerization. Both bonded assemblies were left in this position until complete setting of the resin cement (4 min). The bonded teeth were stored in distilled water at 37 °C for 24 h before further laboratory processing.

TEM examination

A 2 mm thick slab was sectioned from the widest part of each bonded tooth that was designated for TEM examination. These slabs were coated with fast-setting nail varnish applied 1 mm from the bonded interfaces. Without allowing these slabs to be dehydrated, they were immersed immediately in a 50 wt% ammoniacal silver nitrate solution for 24 h, following the protocol for nanoleakage examination described by Tay et al.[10] The silver-stained slabs were rinsed with distilled water and placed in photo-developing solution for 8 h under a fluorescent light to facilitate reduction of the diamine silver ion complexes into metallic silver particles within potential voids along the bonded interfaces. The slabs were dehydrated and embedded in epoxy resin, according to the TEM embedding protocol described by Tay et al.[10] 90–120 nm thick, undemineralised TEM sections were prepared, collected on single slot, carbon- and formvar-coated copper grids (Electron Microscopy Sciences, Fort Washington, PA, USA) and examined without further staining using a transmission electron microscope (Philips 208S, Philips, Eindhoven, The Netherlands) operating at 80 kV.

μTBS evaluation and SEM fractographic analysis

Each tooth was sectioned occluso-gingivally into 0.9 mm thick serial slabs using an Isomet saw under water cooling. Two of these slabs from each tooth were further sectioned into $0.9 \times 0.9\ mm^2$ composite overlay-dentine beams, according to the technique for the 'non-trimming' version of the microtensile test.[15] The exact dimensions of the beams were measured using a pair of digital calipers. The five teeth from each group yielded 31–47 beams for bond strength evaluation. The specimens were stressed to failure under tension using a universal testing machine (Model EMIC DL500, Emic Ltd, S. J. dos Pinhais, PR, Brazil) at a crosshead speed of 0.5 mm/min. The data were analyzed using two-way ANOVA, to examine the effect of materials (i.e. Panavia F vs. Bistite II DC) and application methods (i.e. manufacturer's instructions vs. additional SBMP Plus resin coating), and the interaction of these two factors on bond strength. The total number of tested beams in each group was used in the statistical analysis, with each individual beam considered as an independent specimen. Post hoc comparisons were performed using Student-Newman-Keuls multiple comparison tests at $\alpha = 0.05$.

After the beams were fractured, they were examined using an endodontic microscope (OPMI pico, Carl Zeiss, Oberkochen, Germany) to determine the failure mode. As no gross cohesive failure occurred within the composite overlay or dentine substrate, failure was classified as adhesive failure, along the cement-dentine interface, or mixed failure that occurred both along the interface and within the resin cement. The composite overlay side and the dentine side of representative fractured beams from the four experimental groups were air-dried and sputter-coated with gold/palladium for examination with a scanning electron microscope (Cambridge Stereoscan 360, Cambridge, United Kingdom) operating at 20 kV.

Results

The results of the μTBS tests are shown in Table 2. None of the specimens failed prematurely during sectioning. Two-way ANOVA revealed that neither the factor 'materials' ($P = 0.292$) nor the factor 'application methods' ($P = 0.06$) significantly affected the bond strength results. However, the interaction of these two factors was statistically significant ($P = 0.003$). Student Newman Keul's multiple comparison tests further showed that for Panavia F, there was a statistically significant difference between manufacturer's recommended bonding protocol (i.e. ED primer only) and the additional use of a coating of LVBR ($P < 0.05$). Conversely, there was no difference between the manufacturer's recommended protocol and the use of an additional resin coating for Bistite II DC ($P > 0.05$). When bonding was performed according to the manufacturer's recommended protocol, the mean μTBS of Panavia F was significantly lower than that exhibited by Bistite II DC ($P < 0.05$). By contrast, there was no difference between Panavia F and Bistite II DC when these two resin cements were coupled to primed dentine that were covered with an additional resin coating ($P > 0.05$).

TEM micrographs of dentine coupled with Panavia F are shown in Fig. 1. An overall view of the resin cement revealed the presence of electron-lucent fillers surrounded by a slightly electron-dense coating that were randomly dispersed among the glass fillers within the resin matrix (Fig. 1A). When deep dentine was treated with the ED primer only, the primer could be identified as a 8–9 μm thick, slightly electron-dense layer between the hybrid layer and the resin cement (not shown). In the teeth exposed to silver nitrate, many examined sections revealed large, flat, mushroom-shaped blisters were present within this primer layer that contained extensive silver deposits. Each blister was connected to a patent tubular orifice that also contained silver deposits (Fig. 1B). In addition, resin globules that were devoid of glass filler particles could be identified within the adjacent resin cement matrices (Fig. 1C). These unfilled resin globules were lined with small glass filler particles along their periphery. Some of them were segregated from the rest of the resin matrix and were surrounded by a peripheral rim of silver deposits. However, the majority were well conjugated to the resin cement matrix and could only be identified by the filler particles along their peripheral border, and the presence of isolated silver grains within the resin bodies (Fig. 1C). Both the mushroom-shaped blisters and the resin globules were absent when an additional coat of LVBR was applied, air thinned and light-cured prior to the coupling of the resin cement (Fig. 1D). While the thickness of the hybrid layer remained unchanged, there was a slight increase in the overall thickness of the unfilled resin layer (ca. 15–17 μm thick). The nanoleakage within the hybrid layer was also substantially reduced.

The mushroom-shaped blisters and resin globules were also absent from the interfaces of Bistite II DC when this resin cement was coupled to dentin that was treated with the two-step self-etching primer only (Fig. 2A). However, compared with Panavia F,

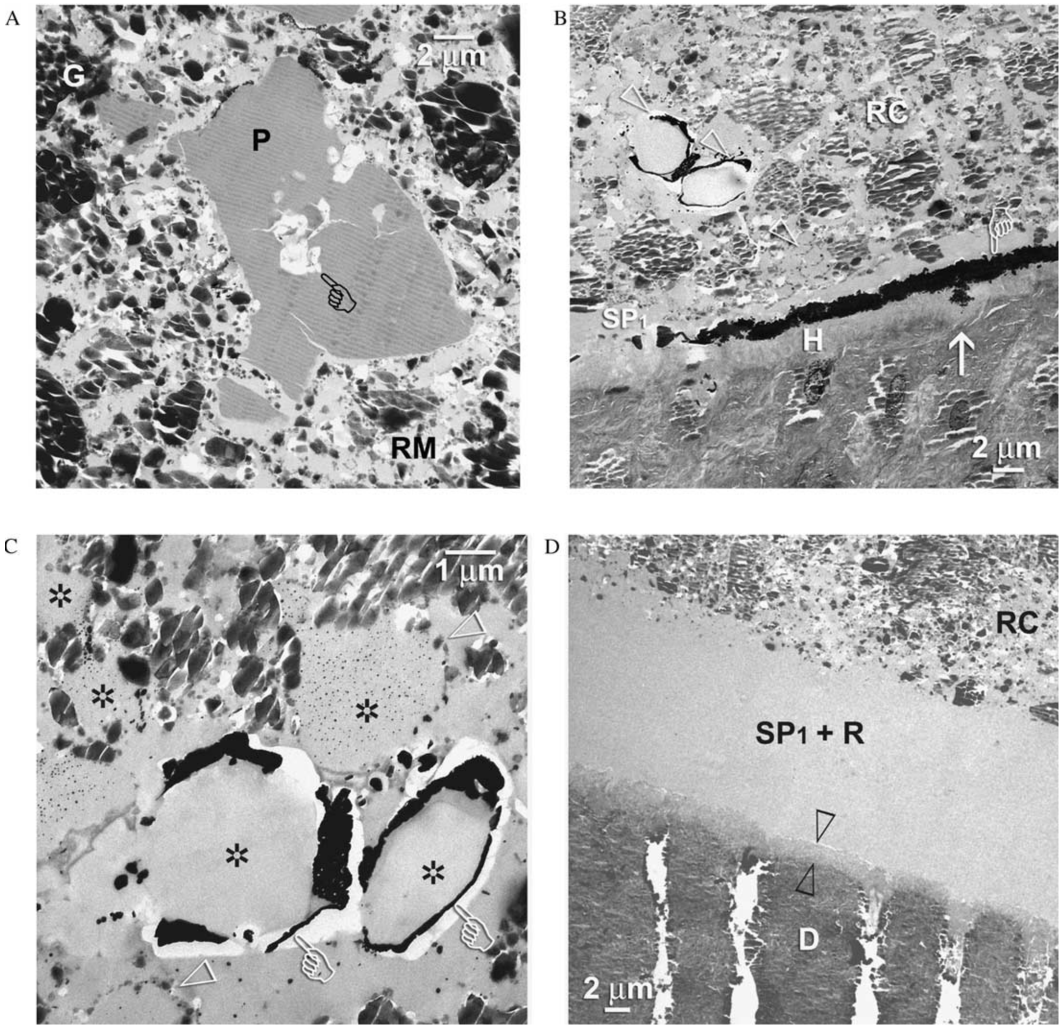

Fig. 1 Unstained TEM micrographs comparing the coupling of Panavia F to dentine with the proprietary one-step, self-etching ED primer only (A–D), or in combination with a light-cured resin coating (E). The specimens were immersed in ammoniacal silver nitrate and the diamine silver ion complexes that penetrated the bonded interfaces were subsequently reduced to metallic silver. (A) An overall view of the resin cement, showing the patented polysiloxane-coated (P) sodium fluoride (pointer) fillers that are incorporated in the resin matrix (RM) for sustained fluoride release. G: barium glass fillers. (B) In sections of deep dentine, there were areas above the hybrid layer (H) in which flat, mushroom-shaped blisters (partially shown) containing extensive silver deposits (pointer) were observed. Each blister was connected to a single, patent dentinal tubule (arrow) that was also filled with silver. Globular unfilled resin bodies (open arrowhead) could also be seen within the matrix of the resin cement (RC). Some of these globules were surrounded by a circumferential layer of silver deposits. They were more easily identified at this magnification. SP_1: ED Primer layer. (C) A high magnification view of the resin globules (asterisks) along the cement–primer junction. These globules were surrounded by a peripheral layer of fine cement particles (open arrowheads). They were probably well conjugated to the resin cement, since there was an absence of circumferential silver uptake. However, isolated silver grains could still be identified within the resin globules. Other globules were surrounded by a peripheral layer of silver deposits (pointer). (D) When SBMP Plus bonding resin was applied after the ED Primer, a 15–17 μm thick electron-lucent layer could be seen ($SP_1 + R$) between the resin cement (RC) and deep dentine (D). The ED primer could not be distinguished from the resin coating. The thickness of the hybrid layer (between arrows) remained unchanged at 2 μm thick. Although nanoleakage could be seen in some parts of the hybrid layer (not shown), mushroom-shaped silver-containing blisters were completely absent.

more extensive nanoleakage was observed in the hybrid layer in Bistite II DC. This resin cement contained glass fillers and highly dispersed, non-agglomerated spherical nanofillers. The resin cement that was close to the primer–cement junction contained isolated silver grains (Fig. 2B) that were absent from the cement that was located further away from this junction. Similar to Panavia F, there was also an increase in the thickness of the primer/bonding resin layer after the use of

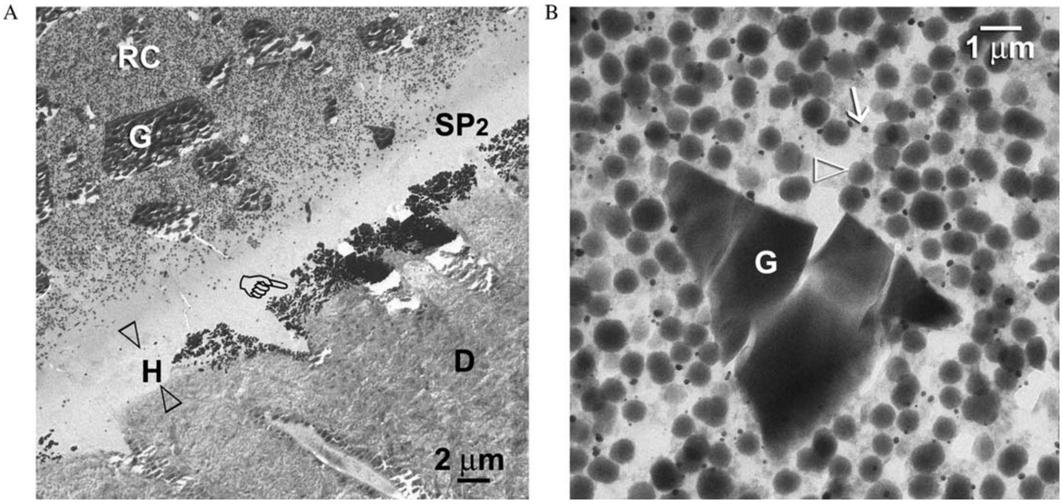

Fig. 2 Unstained TEM micrographs comparing the coupling of Bistite II DC to dentine with the proprietary two-step self-etching primers only (A, B), or in combination with a light-cured resin coating (C). (A) When the two-step, self-etching primers were applied consecutively to deep dentin, a 6–8 μm thick electron-lucent primer layer (SP_2) could be identified between the particulate resin cement (RC) and dentin2 (D). Fairly extensive nanoleakage (pointer) could be observed within the 3 μm thick hybrid layer (H and between arrows). (B) A high magnification view of the resin cement showing the presence of highly dispersed, non-agglomerated spherical zirconia fillers (open arrowhead) among the glass fillers (G). The resin cement that was located close to the cement–primer junction contained additional isolated silver grains (arrow) that were not observed in the resin cement that was located further away from the bonded dentine. (C) When SBMP Plus bonding resin was applied after application of the Bistite II DC primers, a 11–12 μm thick electron-lucent layer could be seen ($SP_2 + R$) between the resin cement (RC) and the deep dentine (D). The primers could not be distinguished from the resin coating. The thickness of the hybrid layer (between arrows) remained unchanged at 3 μm thick. The extent of nanoleakage (pointer) within the hybrid layer (H; between arrows) was also substantially reduced.

an additional coat of LVBR (Fig. 2C). Moreover, less nanoleakage was observed within the hybrid layer following the additional use of the resin coating.

SEM examination of fractured specimen beams revealed that the failure patterns in Panavia F after the use of a resin coating, and Bistite II DC with or without the use of a resin coating were similar (Fig. 3C). However, a unique failure pattern occurred in Panavia F when the dentine was treated only with the ED primer. Numerous circular, rosette-like disks of fractured ED primer, each 15–20 μm in diameter, could be found on both the dentine side (Fig. 3A) and composite overlay side of the fractured beams (not shown). On the dentine side, each rosette exhibited fracture lines that radiated from a patent dentinal tubular orifice (Fig. 3B). In addition, exposed collagen fibrils that were not infiltrated by resin could be identified along the surface of the hybrid layers adjacent to these rosettes.

Discussion

The results demonstrate that the self-etching ED primer permits water-induced interfacial changes that result in lower cement-dentine bond strengths. By covering the primed dentine with a more hydrophobic adhesive layer, these interfacial changes did not occur and the bond strengths increased 35%. We realized that the actual number of teeth employed in each group (vs. number of beams produced from each tooth) was rather small and that inter-tooth differences may exist that affect our conclusion. However, the higher bond strength values obtained from specimens of smaller diameter enable the μTBS test to be discriminative enough for detecting differences arising from treatment variables with the use of a smaller number of actual tooth specimens.

The interfacial structures that were seen in ED primed dentine included unique rosette-like structures seen by SEM. These structures appeared in TEMs as mushroom-like silver deposits. As Bistite II DC already incorporates a two-step self-etching primer, the bond strength of this resin cement was not further improved by the use of an additional coating of LVBR. Nevertheless, severe nanoleakage was observed in this resin cement with the use of the proprietary resin coating (i.e. Primer 2) that consisted of HEMA dissolved in acetone. On the contrary, less nanoleakage was observed in both resin cements with the additional use of the LVBR. Thus, we have to reject the null hypothesis and assert that the use of an additional resin coating

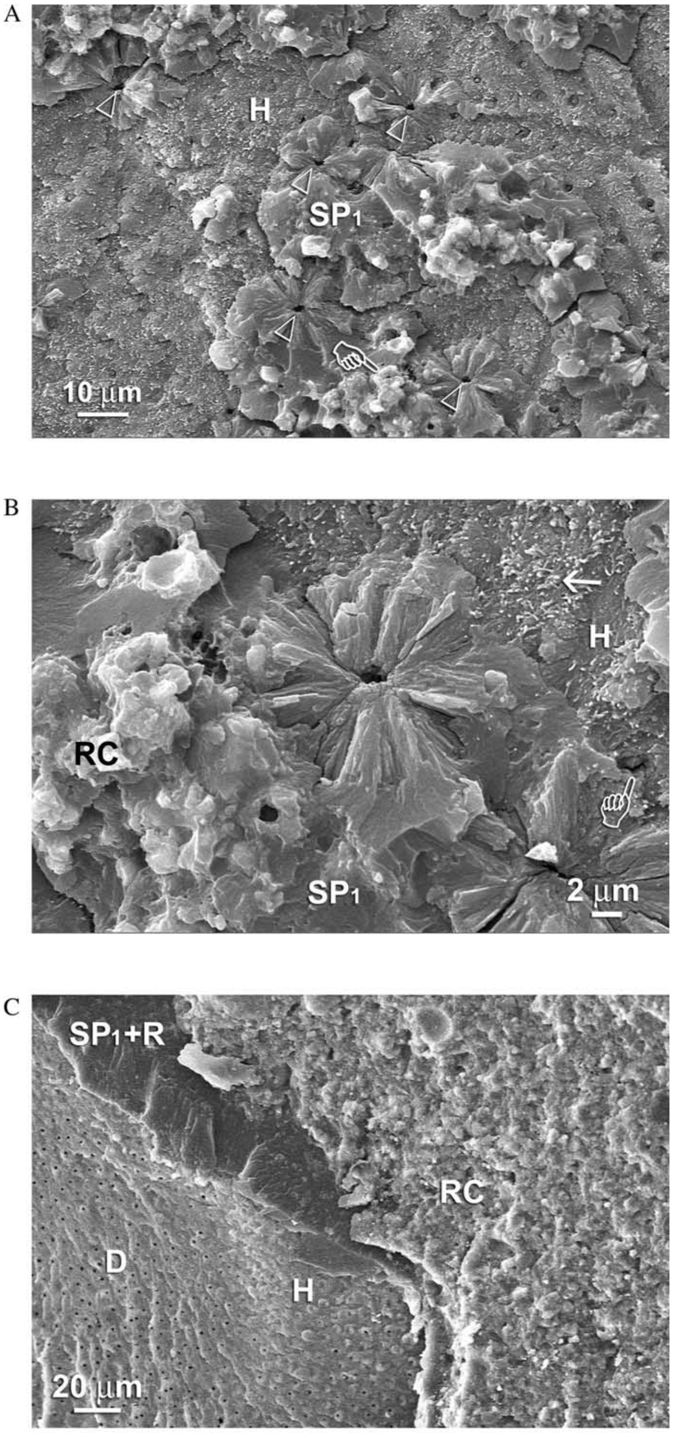

Fig. 3 SEM micrographs of specimen beams examined after tensile testing, depicting unique features that were observed when Panavia F was coupled to dentine that was treated with the one-step ED primer. A. The dentine side of a fractured beam that demonstrated an adhesive failure under optical microscopic examination. SEM revealed a mixed failure mode, with failure occurring predominantly along the surface of the hybrid layer (H), but also cohesively within the resin cement (pointer). The layer of fractured primer (SP_1) exhibited characteristic circular, rosette-like fracture patterns that were around 15–20 μm in diameter (open arrowheads). An empty hole was present in the centre of each rosette. (B) A high magnification view showing that the hole in the centre of a rosette was actually the orifice (pointer) of a patent dentinal tubule. Along the adjacent hybrid layer (H), exposed collagen fibrils (arrows) that were incompletely infiltrated by the ED primer could be seen. SP_1: fractured, unfilled ED primer; RC: fractured particulate resin cement. (C) The rosettes were completely absent when an additional coat of SBMP Plus bonding resin was used with the ED primer (SP_1 + R). A mixed failure was shown in this example. RC: fractured resin cement; H: fractured hybrid layer; D: fractured dentine.

improves the coupling of Panavia F to hydrated dentine.

Improving the adhesion of resin cements to tooth substrates is paramount for increasing the fracture resistance of brittle indirect restorations.[16] Panavia F contains sodium benzene sulphinate in the Primer B component, and a proprietary sodium aromatic sulphinate in the Universal paste of the resin cement to ensure that optimal polymerization of the cement occurs under an acidic environment. It also contains patented polysiloxane-coated sodium fluoride fillers for sustained fluoride release[17] that are probably represented by the coated fillers depicted in Fig. 1A. In the absence of adverse acid–base reactions that resulted in adhesive–composite incompatibility, the low bond strength that was observed when this resin cement was coupled to dentine without a 'resin coating' technique[18] may be explained by the increase in permeability[10] associated with the one-step self-etching ED primer. This increase in adhesive permeability was manifested ultrastructurally by the presence of mushroom-like blisters that were continuous with the lumen of patent dentinal tubules. Presumably, these blisters were either filled with water that permeated from the dentinal tubules, or represented incompletely polymerised regions within the primer layer that resulted from the entrapment of water. When stressed to failure, this extension of the dentinal tubules within the cement may function as sites of stress concentration, resulting in cracks that propagated in a centripetal orientation, and producing the characteristic rosette-like fractures. In addition, the water that permeated through the primer layer may also result in emulsion polymerization of the more hydrophobic resin components within the resin cement, as previously demonstrated by another study.[13] Formation of similar resin globules along the composite–adhesive interface has also been observed when one-step adhesives were employed for the bonding of resin composites.[12]

The TEM observation of fairly extensive nanoleakage within the hybrid layers when both resin cements were used as per the manufacturers' recommendations was also confirmed by the identification of exposed collagen fibrils on the surface of hybrid layers when specimen beams were examined with SEM after tensile testing. These results were contrary to those from a recent micro-Raman spectroscopical study.[19] In that study, the authors claimed that no exposed dentine matrix was present along the dentine/resin cement interface when a self-etching primer was used for conditioning dentine. Conversely, nanoleakage was consistently observed in hybrid layers created by self-etch adhesives.[20,21] In the present study, the application of an additional coating of LVBR appeared to have resulted in, at least qualitatively, a reduction of the nanoleakage within the hybrid layers. As the LVBR is non-solvented and fairly viscous, it is unlikely that nanoleakage could be reduced by the infiltration of this resin into residual spaces within the hybrid layer that were not completely infiltrated by the self-etching primers. It is known that the inclusion of acidic monomers reduces the rate and extent of polymerization of both light- and auto-cured resin blends, with the reduction being more pronounced in the latter.[22] The reduction in the rate of polymerization of self-etching primers may account for the low early bond strengths observed with auto-cured resin cement systems that bond to dentine via a self-etching mechanism.[23] Apparently, early water exposure of self-etching, slow-curing primers or adhesives does compromise their mechanical properties due to plasticisation of the polymer molecules.[24] This renders the adhesive layer weaker and lowers the bond strength. When self-etch adhesives were applied to hydrated dentine and stored for 24 h in water, bond strengths were significantly lower than when similar specimens were stored dry (unpublished observations). The use of an additional light-cured resin coating may have provided additional free radicals to enhance the rate and extent of polymerization of the self-etching primers. This probably reduced the permeability of the adhesive layer to water from the substrate and from the storage media before testing, as shown by the reduced amount of silver impregnation. Reducing the amount of nanoleakage in hybrid layers formed by self-etching primers between resin cements and dentine may improve the long-term durability of these adhesive joints.[25] Further studies should be done to compare the longevity of bonds created by the self-etching type resin cements, with or without the use of a resin coating.

Both the self-etching primers that are used with the resin cements investigated in this study have very low film thickness when they were used according to the manufacturers' instructions (see Figs. 1B, C and 2A). It has also been shown that thin layers of dentine adhesives were less capable of preventing the formation of interfacial gaps.[26] For rigid, non-compliant cast restorations, the contraction stress induced by a resin cement was reported to be inversely proportional to the resin layer thickness.[27,28] Under these situations, even the lower contraction stress of the cements in the auto-cure mode had enough magnitude to disrupt the bonding to dentine.[29] Thus, an additional coat of unfilled resin may also have contributed to the relief of shrinkage stresses in non-compliant adhesive joints.[30] The severe nanoleakage and low film thickness of the primer layer observed when Bistite II DC was used according to the manufacturer's instructions may account for the occasional post-operative sensitivity reported with the use of this resin cement (Junichi Miyata, personal communication). Thus, it would be of clinical interest to see if post-operative sensitivity may be reduced with the use of an additional resin coating prior to the coupling of the resin cement.

Within the limits of this study, it may be concluded that the use of a LVBR is beneficial for the coupling of Panavia F, the resin cement that utilises a one-step self-etching primer to dentine. One additional point of concern is whether the use of such a resin coating will adversely affect the fit of the indirect restorations. Based on the TEM micrographs, it could be seen that provided that the adhesive resin was sufficiently air thinned, the film thickness of the primer layer was increased by no more than 10 μm in both resin cements. The increase in thickness of the primer layer may also be partially compensated by a reduction in the thickness of the resin cement layer. Considering that the cement spaces in indirect ceramic or composite restorations are in the range of 50–100 μm,[31–33] the slight increase in film thickness may not adversely affect the fit of these restorations. Moreover, if impressions are taken after coating the teeth with the additional layer, fitting problems may be eliminated.[14] As the present study was performed on flat dentine surfaces, further studies should be performed to examine the effect of a light-cured unfilled resin coating on the internal adaptation of indirect restorations.

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References

1. Kramer N, Lohbauer U, Frankenberger R. Adhesive luting of indirect restorations. *American Journal of Dentistry* 2000; **13**(Spec No):60D–76D.
2. Rosenstiel SF, Land MF, Crispin BJ. Dental luting agents: a review of the current literature. *Journal of Prosthetic Dentistry* 1998;**80**:280–301.
3. Diaz-Arnold AM, Vargas MA, Haselton DR. Current status of luting agents for fixed prosthodontics. *Journal of Prosthetic Dentistry* 1999;**81**:135–41.
4. Frankenberger R, Kramer N, Petschelt A. Technique sensitivity of dentin bonding: effect of application mistakes on bond strength and marginal adaptation. *Operative Dentistry* 2000;**25**:324–30.
5. Bouillaguet S, Duroux B, Ciucchi B, Sano H. Ability of adhesive systems to seal dentin surfaces: an in vitro study. *Journal of Adhesive Dentistry* 2000;**2**:201–8.
6. Christensen GJ. Solving the frustrations of crown cementation. *Journal of the American Dental Association* 2002;**133**: 1121–2.
7. Van Meerbeek B, De Munck J, Yoshida Y, Inoue S, Vargas M, Vijav P, et al. Buonocore memorial lecture. Adhesion to enamel and dentin: current status and future challenges. *Operative Dentistry* 2003;**28**:215–35.
8. Sensat ML, Brackett WW, Meinberg TA, Beatty MW. Clinical evaluation of two adhesive composite cements for the suppression of dentinal cold sensitivity. *Journal of Prosthetic Dentistry* 2002;**88**:50–3.
9. Sanares AME, King NM, Itthagarun A, Tay FR, Pashley DH. Adverse surface interactions between one-bottle light-cured adhesives and chemical-cured composites. *Dental Materials* 2001;**17**:542–56.
10. Tay FR, Pashley DH, Yiu CKY, Sanares AM, Wei SW. Factors contributing to the incompatibility between simplified-step adhesives and self-cured or dual-cured composites. Part I. Single-step self-etch adhesive. *Journal of Adhesive Dentistry* 2003;**5**:27–40.
11. Ikemura K, Endo T. Effect on adhesion of new polymerization initiator systems comprising 5-monosubstituted barbituric acids, aromatic sulphonate amides, and *tert*-butyl peroxymaleic acid in dental adhesive resin. *Journal of Applied Polymer Science* 1999;**72**:1655–68.
12. Tay FR, Pashley DH, Suh BI, Carvalho RM, Itthagarun A. Single-step adhesives are permeable membranes. *Journal of Dentistry* 2002;**30**:371–82.
13. Mak YF, Lai SCN, Cheung GSP, Chan AW, Tay FR, Pashley DH. Micro-tensile bond testing of resin cements to dentin and an indirect resin composite. *Dental Materials* 2002; **18**:609–21.
14. Jayasooriya PR, Pereira PN, Nikaido T, Barrow MF, Tagami J. The effect of a 'resin coating' on the interfacial adaptation of composite inlays. *Operative Dentistry* 2003; **28**:28–35.
15. Shono Y, Ogawa T, Terashita M, Carvalho RM, Pashley EL, Pashley DH. Regional measurement of resin–dentin bonding as an array. *Journal of Dental Research* 1999;**78**:699–705.
16. Furukawa K, Inai N, Tagami J. The effects of luting resin bond to dentin on the strength of dentin supported by indirect resin composite. *Dental Materials* 2002;**18**: 136–42.
17. Kawashima M. Kenichi H. Coated metal fluoride particles and a dental composition containing coated metal fluoride particles. United States Patent number 5,908,879; 1999.
18. Jayasooriya PR, Pereira PN, Nikaido T, Tagami J. Efficacy of a resin coating on bond strengths of resin cement to dentin. *Journal of Esthetic and Restorative Dentistry* 2003;**15**: 105–13.
19. Walker MP, Wang Y, Spencer P. Morphological and chemical characterization of the dentin/resin cement interface produced with a self-etching primer. *Journal of Adhesive Dentistry* 2002;**4**:181–9.

20. Li H, Burrow MF, Tyas MJ. The effect of concentration and pH of silver nitrate solution on nanoleakage. *Journal of Adhesive Dentistry* 2003;**5**:19–25.
21. Tay FR, King NM, Chan KM, Pashley DH. How can nanoleakage occur in self-etching adhesive systems that demineralize and infiltrate simultaneously? *Journal of Adhesive Dentistry* 2002;**4**:255–69.
22. Tay FR, Feng L, Suh BI, Pashley DH. Factors contributing to the incompatibility between simplified-step adhesives and chemical-cured or dual-cured composites. Part III. Effect of acidic resin monomers. *Journal of Adhesive Dentistry* 2003; in press.
23. Burrow MF, Nikaido T, Satoh M, Tagami J. Early bonding of resin cements to dentin—effect of bonding environment. *Operative Dentistry* 1996;**21**:196–202.
24. Carrilho MRO, Carvalho RM, Tay FR, Pashley DH. Effects of storage medium on the mechanical properties of adhesive systems. *American Journal of Dentistry* 2003; in press.
25. Kitasako Y, Burrow MF, Katahira N, Nikaido T, Tagami J. Shear bond strengths of three resin cements to dentine over 3 years in vitro. *Journal of Dentistry* 2001;**29**:139–44.
26. Koike T, Hasegawa T, Itoh K, Yukitani W, Yamashita T, Wakumoto S, et al. Effect of multiple application of a dentin adhesive on contraction gap width of a resin-based composite. *American Journal of Dentistry* 2002;**15**:159–63.
27. Alster D, Feilzer AJ, De Gee AJ, Davidson CL. Tensile strength of thin resin composite layers as a function of layer thickness. *Journal of Dental Research* 1995;**74**: 1745–8.
28. Alster D, Feilzer AJ, De Gee AJ, Davidson CL. Polymerization contraction stress in thin resin composite layers as a function of layer thickness. *Dental Materials* 1997;**13**: 146–50.
29. Braga RR, Ferracane JL, Condon JR. Polymerization contraction stress in dual-cure cements and its effect on interfacial integrity of bonded inlays. *Journal of Dentistry* 2002;**30**:333–40.
30. Choi KK, Condon JR, Ferracane JL. The effects of adhesive thickness on polymerization contraction stress of composite. *Journal of Dental Research* 2000;**79**:812–7.
31. Audenino G, Bresciano ME, Bassi F, Carossa S. In vitro evaluation of fit of adhesively luted ceramic inlays. *International Journal of Prosthodontics* 1999;**12**:342–7.
32. Molin MK, Karlsson SL, Kristiansen MS. Influence of film thickness on joint bend strength of a ceramic/resin composite joint. *Dental Materials* 1996;**12**:245–9.
33. Wassell RW, Gagliano G. Effects of adhesive fixed prosthesis retainer design on resultant resin luting agent thickness. *Journal of Prosthetic Dentistry* 1998;**80**:479–84.