Dental Materials (2006) **22**, 45–56

dental materials

www.intl.elsevierhealth.com/journals/dema

Micro-tensile bond strength of three luting resins to human regional dentin

Bin Yang[a], Klaus Ludwig[a], Rainer Adelung[b], Matthias Kern[a,*]

[a]*Department of Prosthodontics, Propaedeutics and Dental Materials, Christian-Albrechts University, Arnold-Heller-Strasse 16, 24105 Kiel, Germany*
[b]*Multicomponent Materials, Faculty of Engineering, Christian-Albrechts University, Kiel, Germany*

Received 13 July 2004; received in revised form 28 January 2005; accepted 8 February 2005

KEYWORDS
Micro-tensile bond;
Regional dentin;
Luting resin;
TEM;
SEM;
Interface;
Fractography

Summary *Objectives*: To evaluate the micro-tensile bonding strength (μTBS) of three luting resins to human regional dentin.
Methods: Dentin disks from non-carious third molars were prepared from different regions (s, superficial dentin; d, deep dentin; c, cervical dentin), and divided into groups based on anatomical locations and luting resins (Super-Bond C&B: SB; Panavia F 2.0: PF; RelyX Unicem: RU): SB-s, SB-d, SB-c; PF-s, PF-d, PF-c; RU-s, RU-d, RU-c. Luting resins were used according to the manufacturers' instructions, to bond 1-mm-diameter PMMA or composite rods to the exposed dentin specimens under a load of 7.5 N, in the self-curing mode. After storage for 1 or 3 days, μTBS was tested at a cross-head speed of 1 mm/min. Data were analyzed with ANOVA and Fisher's PLSD test. The bonding interface and fractography analyses were performed with SEM and TEM.
Results: ANOVA results showed that μTBS to superficial dentin was significantly higher than to deep or cervical dentin for all three luting resins. SB-s and PF-s, with the highest μTBS, failed primarily cohesively in luting resin. μTBS of SB-d and SB-c were significantly higher than those of PF and RU. RU, with the lowest regional μTBS, failed mostly within demineralized dentin. SEM and TEM showed that adhesive failures in SB and PF occurred at the top of the hybrid layer (HL), but no obvious HL was observed in RU.
Significance: Luting resins with different chemical formulations and applications yield significantly different bond strengths to different regions in human dentin.

Introduction

Resin cements are increasingly used for luting all-ceramic, metal or composite indirect restorations due to their excellent mechanical properties, better bond strengths and improved esthetics

* Corresponding author. Tel.: +49 431 5972874; fax: +49 431 5972860.
E-mail address: mkern@proth.uni-kiel.de (M. Kern).

0109-5641/$ - see front matter © 2005 Academy of Dental Materials. Published by Elsevier Ltd. All rights reserved.
doi:10.1016/j.dental.2005.02.009

when compared to conventional cements [1]. With the growing understanding of dentin and dentin smear layers, it is now recognized that the smear layer should be removed or modified and the underlying dentin should be demineralized to expose the three-dimensional collagen network that can be infiltrated by adhesive resin monomers [2,3] to form a hybrid layer (HL) between luting resins and dentin. Dentin is a hydrated composite material composed of the collagen-based organic matrix with mineral reinforcement, varying with anatomical location [4]. The structural anisotropy in regional dentin responds differently to etching and priming, or self-etching primers/adhesives, during dentin bonding procedures, and consequently, the conditioned dentin shows varying permeability to luting resins and hence, varying bond strengths [3]. In general, bond strengths are higher in superficial dentin than in deep dentin [5,6]. Burrow [7] suggested that bond strength was related more to the quality of the HL than to the depth of dentin etching. However, resin bonding of the cervical margin was less predictable due to the oblique tubule orientation [8] and the lower density of tubules than in deep dentin [9].

In order to simplify application procedures, and to prevent the collapse of the collagen fibril network of demineralized dentin, two-step self-etching primer systems and one-step self-etching adhesive systems have been developed in recent years. However, the literature has reported conflicting results on bond strengths of self-etching systems to dentin [10], and some recent studies suggested that combining the primer and adhesive resins into a single application step may reduce the quality of the hybridization of dentin [11,12]. Little information is available about the bond strength of self-etching luting resins to different regions of dentin and their bonding mechanism.

Recently, the micro-tensile bond strength test (μTBS) [13,14] has become popular for testing adhesion to dentin because this technique, presumably, provides better stress distribution at the adhesive interface due to the small bonding area, with fewer defects than in standard tensile tests. Also, this technique can be used to detect regional difference in resin–dentin bond strengths due to its use of small bonding areas [6].

The aim of the present study was to evaluate SEM and TEM ultrastructures and μTBS of three luting resins, used in their self-curing modes, to different regions of dentin. The null hypotheses that were tested were (1) μTBS of three luting resins to dentin do not vary with dentin location; (2) the different chemical formulations of the three luting resins and their application instructions do not result in different morphological appearances of the bonding interfaces and failure modes.

Materials and methods

Tooth preparation

Intact caries-free human molars extracted from individuals 18–45 years old were stored in 0.5% chloramine T solution for 2 weeks, then in distilled water at 4 °C prior to preparation. The teeth were used within 3 months after extraction. In this study, the age difference among the collected teeth was ignored since a previous study showed that age did not greatly influence the dentin bond strength [7]. Dentin disks (about 1.5-mm thick) were prepared by cutting occlusal enamel and dentin perpendicular to the tooth axis 1 mm below the dentino-enamel junction (DEJ) (s, superficial dentin), 1 mm above the pulp horn (d, deep dentin), or parallel to the tooth axis, 0.5 mm above the cemento-enamel junction (CEJ) and 0.5 mm below the DEJ (c, cervical dentin) using a slow-speed saw with a diamond-coated disk (Isomet, Buehler, Lake Bluff, IL, USA) under water cooling (Fig. 1a). From each molar, 2–4 superficial dentin disks and two cervical dentin disks, or 2–4 deep dentin disks could be obtained. Then dentin specimens were wet polished with 600 grit SiC paper and stored in distilled water at 4 °C. The dentin specimens from each region were randomly divided into the test groups for bonding.

Micro-tensile bond strength (μTBS) testing

Super-Bond C&B unfilled luting resin (SB; Sun Medical, Shiga, Japan), Panavia F 2.0 composite luting resin combined with ED self-etching primer 2.0 (PF; Kuraray Medical Inc., Osaka, Japan), and RelyX Unicem self-etching adhesive luting resin (RU; 3M Espe AG, Seefeld, Germany) were used for bonding. The luting resins were used in the self-curing mode according to manufacturers' instructions (Table 1). Ten-micrometer-thick aluminum foil with a 1-mm-diameter hole was attached to each conditioned or non-conditioned dentin surface. The hole was located at the center of the bonding area using an alignment jig (Fig. 1b). In order to obtain a reliable bond between the handling rod with a diameter of 2 mm and luting resin, PMMA rods for SB or composite rods made of Clearfil FII composite resin (Kuraray Medical Inc., Osaka, Japan) for PF

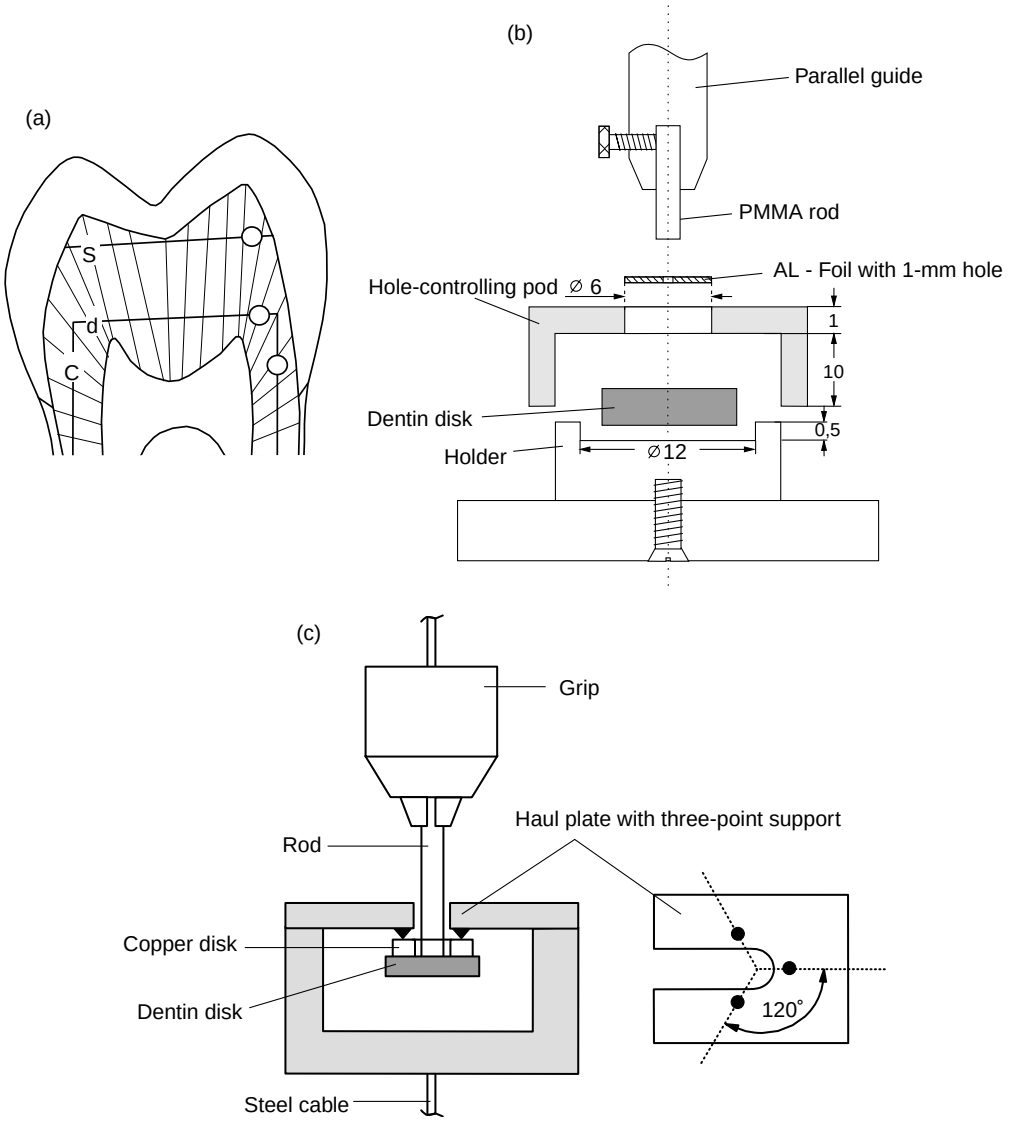

Figure 1 (a) Diagram of tested dentin location. s, superficial dentin (1 mm below DEJ); d, deep dentin (1 mm above the pulp horn); c, cervical dentin (0.5 mm below DEJ, 0.5 above CEJ); O, tested dentin location. (b) Alignment apparatus. Ten-micrometer-thick aluminum foil with a 1-mm-diameter hole was attached to dentin surface. The hole was located at the center of the bonding area using an alignment jig to control the shape and size of bonding area. (c) Schematic drawing of μTBS testing. The rod was gripped by a pin-vice of a universal testing machine, and then a haul plate with three point support was put on the dentin surface. The μTBS was measured only by tensile force.

and RU were bonded perpendicularly with the luting resins on the exposed dentin surface under a load of 7.5 N. After 37 °C water storage for 24 h (groups SB and PF) or 37 °C at 100% relative humidity (RH) for 72 h (group RU), μTBS testing was performed with a universal testing machine (Zwick Z010/024, Zwick, Germany) at a cross-head speed of 1 mm/min. The PMMA or composite rods were gripped in a pin-vice (Fig. 1c). Based on the luting resins and dentin regions, the test groups with 12 specimens each were classified into: SB-s, SB-d, SB-c; PF-s, PF-d, PF-c; RU-s, RU-d, RU-c. In each group, eight bonded specimens were used for μTBS testing, and four specimens for TEM examination.

SEM examination and fractography analysis

Dentin specimens acid-etched using SB green activator (10% citric acid with 3% ferric chloride: 10–3 solution) for 10 s or self-etching ED primer 2.0 for 30 s, were fixed with 2% glutaraldehyde in phosphate buffer for 8 h, then dehydrated in an ascending ethanol series (50, 60, 70, 80, 90, 96 and 100%) for 1 h each. After the critical point drying procedure (K850 Critical Point Dryer, Emitech Ltd,

Table 1 Composition and application of the test luting resins (batch number in parenthesis).

Adhesive luting resin	Components	Etching	Priming	Bonding procedures	Storage condition
Super-Bond C&B (SB)	**Green activator** (EM 1)	Etch dentin with green activator for 10 s, rinse and air dry gently	Prewet dentin with 4META/MMA-TBB	Mix liquid and powder with brush-on technique. Apply to dentin surface. Bond the PMMA rod to dentin surface. Place the bonded specimen at room temperature for 6 min	24 h in 37 °C water
Self-curing unfilled luting resin	**Monomer** (FG 2)				
Sun Medical Co. Ltd, Shiga, Japan	**Polymer** L-type radiopaque (FE 2) **Catalyst S** (EM 12)				
Self-etching primer (PF)	**ED Primer 2.0 A** (00161A)	Treat dentin with self-eching ED primer 2.0 for 30 s, air dry gently		Mix Panavia F 2.0, apply to the composite rod, then bonded to treated dentin. After removal of excess resin, Oxyguard II 2.0 applied to the luting margins. Placed into 37 °C incubator for 20 min	24 h in 37 °C water
Kuraray Medical Inc., Osaka, Japan	**ED Primer 2.0 B** (00044)				
Panavia F 2.0 (PF) Two-step self-etching luting resin	**A paste** (0001A)				
Kuraray Medical Inc., Osaka, Japan	**B paste** (00001A)				
RelyX Unicem (RU)	**Aplicap**	None	None	Mix RelyX Unicem, apply to the composite rod, then bond to dentin without treating dentin at room temperature for 30 min	72 h in 100% RH at 37 °C[a]
One-step self-etching luting resin	**Self-adhesive universal resin cement**				
3M ESPE AG Seefeld, Germany	(152009)				

[a] Recommended by 3M ESPE company, 72 h in 100% RH at 37 °C was used for the complete curing of RelyX Unicem luting resin in self-curing mode.

UK), the specimens were gold-sputtered and examined by a scanning electron microscope (SEM, Philips XL 30 CP, Philips, Germany) operating at 10–25 kV.

After μTBS testing, the debonded dentin specimens were air-dried for 24 h, gold-sputtered and observed by SEM to evaluate the failure modes. Failure modes were classified into one of the following modes: (A) adhesive failure along dentin surface; (B) mixed failure: adhesive failure with a thin layer of luting resin remaining on the dentin surface; (C) cohesive failure in luting resin. The fractured area of each failure mode on the dentin surfaces was determined from the SEM micrographs with scale paper and expressed as a percentage of the total bonding surface area for each test group.

Statistics analysis

The data of μTBS of the three luting resins to regional dentin were statistically analyzed with a two-way ANOVA (materials vs. region) and Fisher's PLSD test at a confidence level of 95%. The failure mode results were compared for each luting material using the Mann–Whitney *U*-test.

TEM examination

After 24 (SB and PF) or 72 h (RU) of water storage, bonded and debonded dentin specimens were immediately immersed into 2.5% glutaraldehyde in phosphate buffer solution for 4 h. After fixation, the specimens were demineralized in 4% EDTA buffered to pH 7 for 7 days, postfixed with 1% osmium tetroxide for 2 h, and then dehydrated in an ascending ethanol series (30, 40, 50, 60, 70, 80, 90, 96 and 100%) twice in each solution for 10 min each time. Finally, the dehydrated specimens were embedded in pure epoxy resin (Araldite CY212, 13824, Serva, Germany) in a 60 °C oven for 48 h. Semi-thin sections of about 70-nm thick were prepared with an ultramicrotome (Reichert Ultracut E, Leica, Austria) and stained with saturated uranyl acetate for 10 min and lead citrate for 5 min, and examined with a transmission electron microscope (TEM 201, Phillips, The Netherlands).

Results

Means and SDs of μTBS of the various dentin regions of the three luting resins are shown in Table 2. Two-way ANOVA revealed that both the factors tested (luting resin and regional location) and their interaction had significant influences on μTBS. Fisher's PLSD multiple comparison tests further showed that for all three luting resins, the mean μTBSs to superficial dentin were significantly higher than those to deep or cervical dentin ($p \leq 0.05$). There were no significant differences in μTBS between deep dentin and cervical dentin groups. The μTBS of SB-s and PF-s groups was significantly higher than that of group RU-s ($p \leq 0.05$), whereas no difference was detected between SB-s and PF-s. In deep and cervical dentin, the μTBS of SB was significantly higher than those of PF and RU with the lowest μTBS seen in group RU ($p \leq 0.01$). The μTBS of specimens luted with RU was significantly lower in all regions than those of the other two luting resins ($p \leq 0.01$). No premature bond failures occurred during the μTBS testing in any of groups.

The failure modes of the three luting resins during μTBS testing are shown in Fig. 2. Statistically significant differences were found among regional dentin sites for SB and PF ($p \leq 0.01$), and for RU ($p \leq 0.05$). For group SB-s, failures were mostly cohesive (68%) in the luting resin. In deep and cervical dentin, most of the failures were observed to be adhesive failures along the dentin surface for SB-d (74%) and SB-c (45%). For groups PF, 46% of failures occurred cohesively in luting resin in superficial dentin while failures in deep dentin were mostly adhesive in nature (76%). In contrast, for groups RU, most of the failures to regional dentin were found to be adhesive along the dentin surface or partially adhesive failures with a thin layer of cohesively fractured luting resin. No adhesive failures were seen between PMMA rod-cement or composite rod-cement interfaces and no cohesive failures in the demineralized dentin under the HL were observed in any of μTBS test specimens.

The SEM observations of the etched treated dentin in specimens from the SB and PF groups are shown in Fig. 3a and b. After the dentin was etched with 10-3 solution (SB), the smear plugs appeared to be removed and the tubule orifices were completely exposed (Fig. 3a). Some residual smear layer material was seen around tubule orifices. Circumferentially oriented collagen fibrils around the tubule wall were exposed. After the dentin was treated with self-etching ED primer 2.0 (Fig. 3b), the smear layer appeared to be demineralized, exposing collagen fibrils on the intertubular dentin surface. Some smear plugs were only partially removed leaving some smear debris in the tubules.

Table 2 Micro-tensile bond strength (μTBS) of the test groups to human regional dentin.

Groups	Super Bond C&B	Panavia F 2.0	RelyX Unicem
Superficial dentin	$31.9(7.2)^{A}_{a}$	$29.1(8.4)^{A}_{a}$	$8.2(2.5)^{A}_{b}$
Deep dentin	$18.6(4.3)^{B}_{a}$	$10.4(1.9)^{B}_{b}$	$5.7(2.0)^{B}_{c}$
Cervical dentin	$24.2(6.5)^{B}_{a}$	$10.2(3.6)^{B}_{b}$	$5.5(2.0)^{B}_{c}$

Means (SD) in MPa. Within the same column means with the same upper case superscript letter are not statistically different ($p > 0.05$). Within the same row means with the same lower case subscript letter are not statistically different ($p > 0.05$). Two-way ANOVA followed by Fisher's PLSD multiple comparison tests results at a confidence level of 95%. $N = 8$.

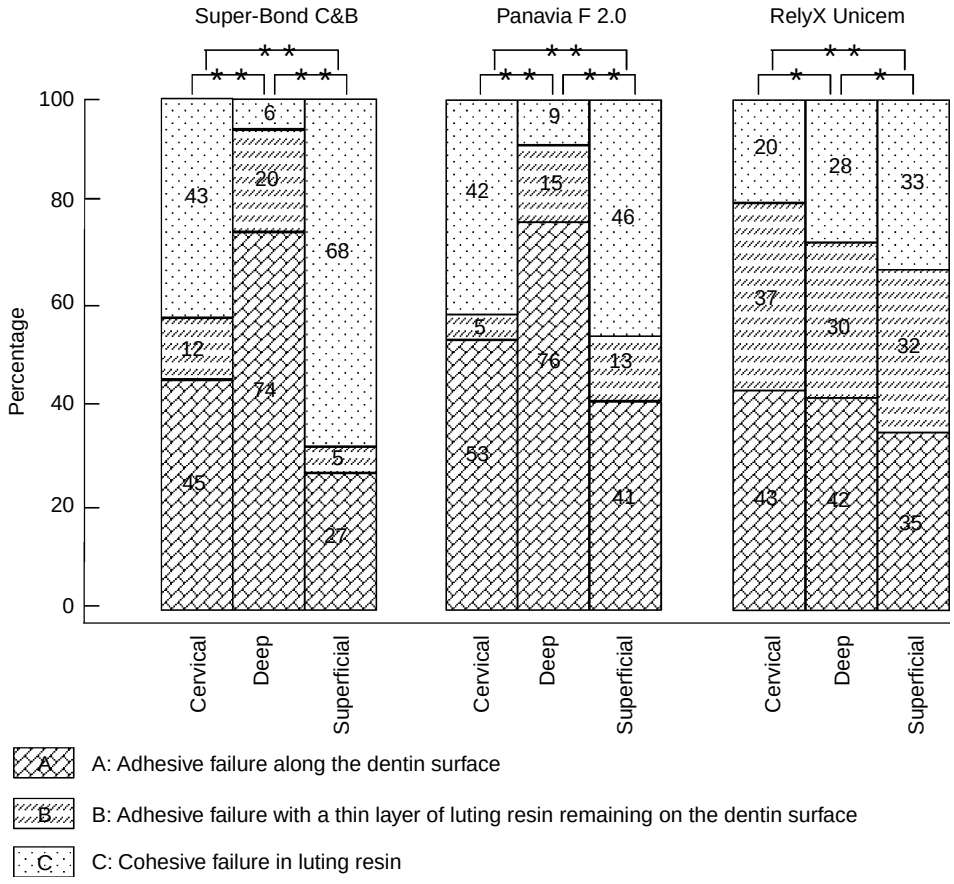

Figure 2 Failure modes of Super-Bond C&B, Panavia F 2.0 and RelyX Unicem to dentin regions during the tensile bonding strength test. Horizonal lines indicate that tested groups at both ends are statistically different (Mann–Whitney *U*-test) (**$p \leq 0.01$, *$p \leq 0.05$).

Some peritubular dentin remained in PF specimens. The polished and untreated dentin used in the RU group was covered with a smear layer (Fig. 3c).

Figs. 4–6 present examples of the interface and fractured surfaces of dentin bonded with three luting resins using SEM and TEM. In group SB specimens, a HL with a width of approximately 4 μm was formed (Fig. 4a) between the SB luting resin and superficial dentin. Adhesive failure occurred along the top of HL on deep dentin surfaces with cohesive failure within the resin tags (Fig. 4b and c). The hybrid layer on the deep intertubular dentin surface extended into the tubule walls surrounding the resin tags, occluding the tubule openings (Fig. 4d). In group PF specimens, the HL between PF luting resin and superficial dentin was approximately 1.5–2 μm thick, consisting of a 0.5 μm hybridized smear layer and a 1–1.5 μm thick authentic HL (Fig. 5a). Adhesive failure was found at the top of HL on deep dentin with cohesively fractured resin tags occluding the tubules (Fig. 5b and c). A tubule cut obliquely revealed the presence of a lining membrane within the tubule orifice (Fig. 5d). In group RU specimens, no obvious HL was observed (Fig. 6a). Adhesive failure occurred at the top of the demineralized deep dentin surface with cohesively fractured resin tags occluding the tubules (Fig. 6b and c). Loose collagen fibrils on the dentin surface of a adhesively debonded cervical dentin specimen do not seem to be enveloped by the luting resin (Fig. 6d).

Discussion

In order to obtain reliable initial tensile bond strengths of the three luting resins to dentin in self-curing mode, two different storage conditions, i.e. 24 and 72 h, respectively, were used separately in the test groups, according to the manufacturers' suggestions. The curing degree of resins is an important factor influencing the bond strength. For SB and PF, the radical polymerization reaction should be almost completed and stable after 24 h water storage. For RU, the setting reaction is completed only after 72 h at 100% RH, because

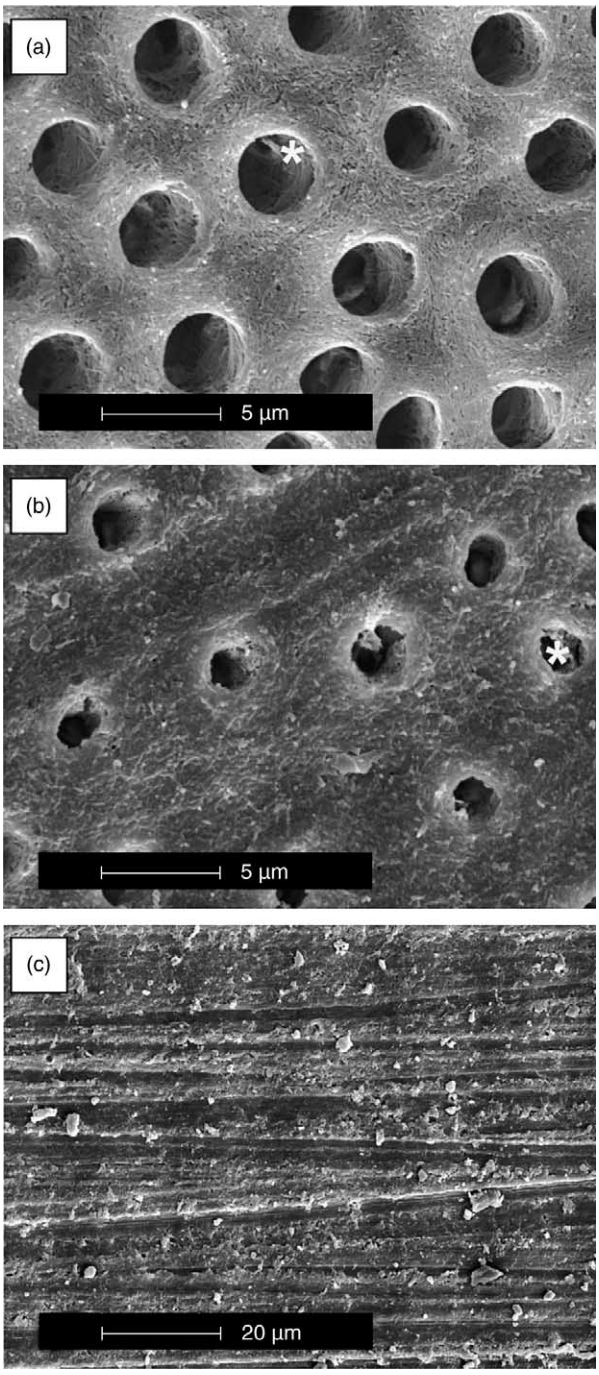

Figure 3 SEM micrographs of conditioned and unconditioned dentin surfaces, as the bonding substrates in the three groups. (a) SEM micrograph at 3400 magnification illustrating the dentin surface etched with 10% citric acid with 3% ferric chloride (group Super-Bond C&B). The smear plugs appeared to be removed and the tubule orifices were exposed completely by removal of peritubular dentin. Some residual smear layer material was seen around tubule orifices. The circumferentially oriented collagen fibrils (asterisk) around the tubular wall are exposed. (b) SEM micrograph at 3400 magnification illustrating the dentin surface treated with self-etching ED primer 2.0 (group Panavia F 2.0). The dentin surface exhibited demineralized collagen fibrils. The tubule orifices were exposed with some smear debris (asterisk) remaining in the tubules. (c) SEM micrograph at 650 magnification of polished and unconditioned dentin surface covered by a smear layer (group RelyX Unicem).

the cement reaction between the acidic methacrylate and basic fillers is also included in the setting reaction, apart from the radical polymerization reaction (Technical Product Profile of RelyX Unicem Aplicap/Maxicap Self-Adhesive Universal Resin Cement). Therefore, although the storage conditions for the three luting resins are different, it is thought that this difference did not significantly influence the bond strength testing especially as the bond strengths were not improved in group RU, even after exclusion from water for 72 h.

The μTBS of SB-s and PF-s in the present study was 31.9 and 29.1 MPa, respectively. This exceeds the results from a previous study [15] in which the μTBS of Super-Bond C&B and Panavia F exhibited 24.7 and 16.1 MPa. In that study, composite overlays were bonded to flat deep dentin surfaces polished with 180 grit SiC paper, followed by cutting the bonded specimens into 0.9×0.9 mm composite-dentin beams. However, the thickness and quality of smear layer produced by 180 grit SiC paper and their influence on bond strength are significantly different from those produced by 600 grit in this study [16,17].

The configuration factor (C-factor) has been accepted as an important factor influencing bond strength during bonding procedure [18]. Bouillaguet [19] found that the μTBS of SB and PF to flat root dentin with a low C-factor were significantly higher than those to intact root canal dentin with a high C-factor. In the current study, flat dentin disks with a low C-factor were used for bonding, instead of crown segment containing tooth preparations with a relatively high C-factor. Therefore, differences in smear layer, geometry of bonding area [20], C-factor and regional difference of dentin substrates might account for the different bond strengths in the current study compared to the cited studies.

In the present study, μTBSs to superficial dentin were significantly higher than those to deep dentin and cervical dentin for all luting resins, which is in good agreement with previous studies [6]. In superficial dentin there is more intertubular dentin area rich in collagen fibrils than in deep and cervical dentin. Therefore, the μTBS was significantly higher in superficial dentin due to the opportunity for more micromechanical adhesion to collagen fibrils in the HL [2–4]. In group SB-s, the mean μTBS of 31.9 MPa

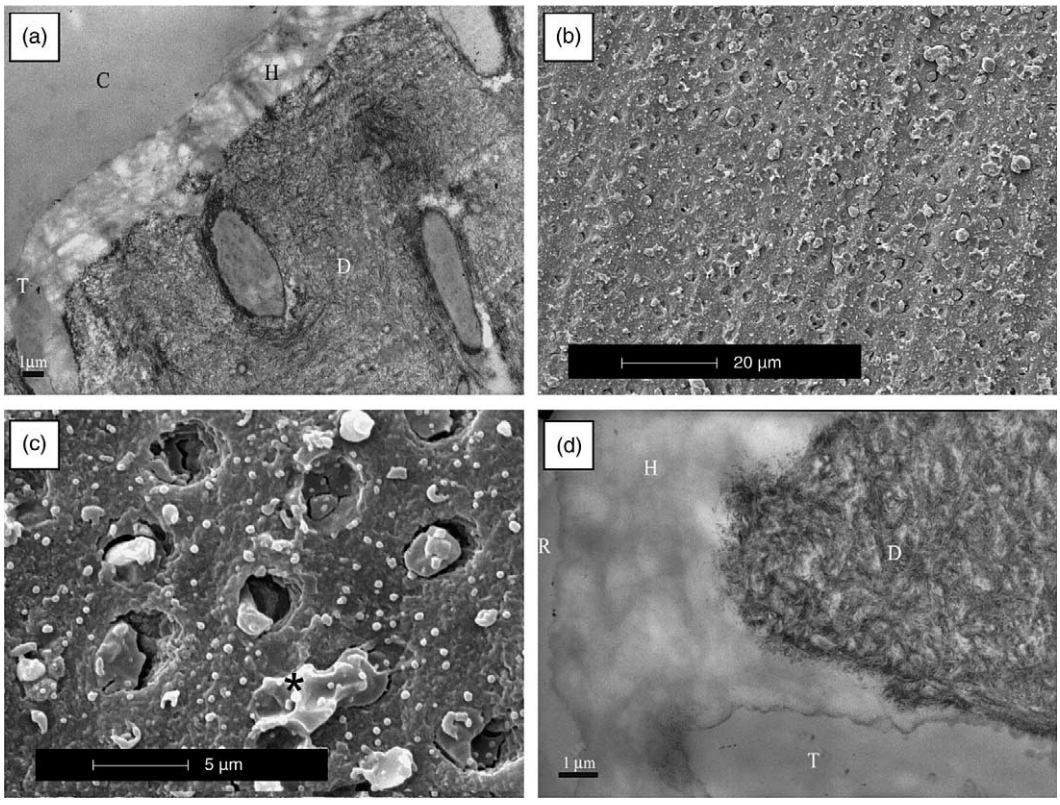

Figure 4 Interface and fractography analysis of group SB (Super-Bond C&B). (a) TEM photomicrograph at 3000 magnification illustrating an overview of the interface between SB luting resin and superficial dentin. The hybrid layer is approximately 4 μm thick. (b) SEM micrograph at 600 magnification of a debonded deep dentin specimen where adhesive failures occurred on the dentin surface. (c) SEM micrograph at 3600 magnification of the same specimen as in (b). Adhesive failure occurred along the top of the hybrid layer and cohesive failures occurred in the resin tags (asterisk). (d) TEM micrograph at 6000 magnification of a resin tag in deep dentin. The hybrid layer on the intertubular dentin surface extended into the tubule walls surrounding the resin tag, occluding the tubule opening. C, luting resin; H, hybrid layer; T, resin tag; D, demineralized dentin in the preparation of TEM; R, embedding resin.

and the high percentage of cohesive failures (68%) in bulk luting resin demonstrated that the initial μTBS to superficial dentin was higher than the cohesive strength of the luting resin.

For group PF-s, the mean μTBS of 29.1 MPa and 46% of cohesive failure in luting resin also demonstrated that the bonded interface was stronger compared to the cohesive strength of luting resin. This shows that the ED primer 2.0 successfully etched through the smear layer to partially demineralize the underlying dentin and improved the permeability of dentin to resin monomers (Figs. 3 and 5). Therefore, the smear layer and partially demineralized dentin could be incorporated into a hybridized complex by infiltration and polymerization of resin monomers. However, the top of the hybridized smear layer appears to be a potential weak link as the smear layer is weaker than sound dentin [2], since there was a high percentage of adhesive failure at the top of HL (Fig. 2).

In the present study, the μTBS of RU to different dentin regions was significantly lower than those of the other two luting resins, although its μTBS to superficial dentin was statistically higher than to deep and cervical dentin. Theoretically, the acidic polymerizable methacylate-based monomers in RelyX Unicem, a bis-GMA/TEDGMA based resin, typically have at least two phosphoric acid groups and a minimum of two C=C double bond units per molecule. With the presence of water, these monomers should demineralize the smear layer and the underlying dentin and simultaneously infiltrate the porous dentin surface due to their hydrophilic properties [21]. However, the TEM micrograph showed that no obvious HL was formed at the resin–dentin bonding interface (Fig. 6). Resin infiltration is proportional to the applied concentration, viscosity of the solution, molecular weight or size, the affinity of monomers for the substrate and the time allowed for penetration [2]. RelyX Unicem is a heavily filled (72 wt% reactive glass

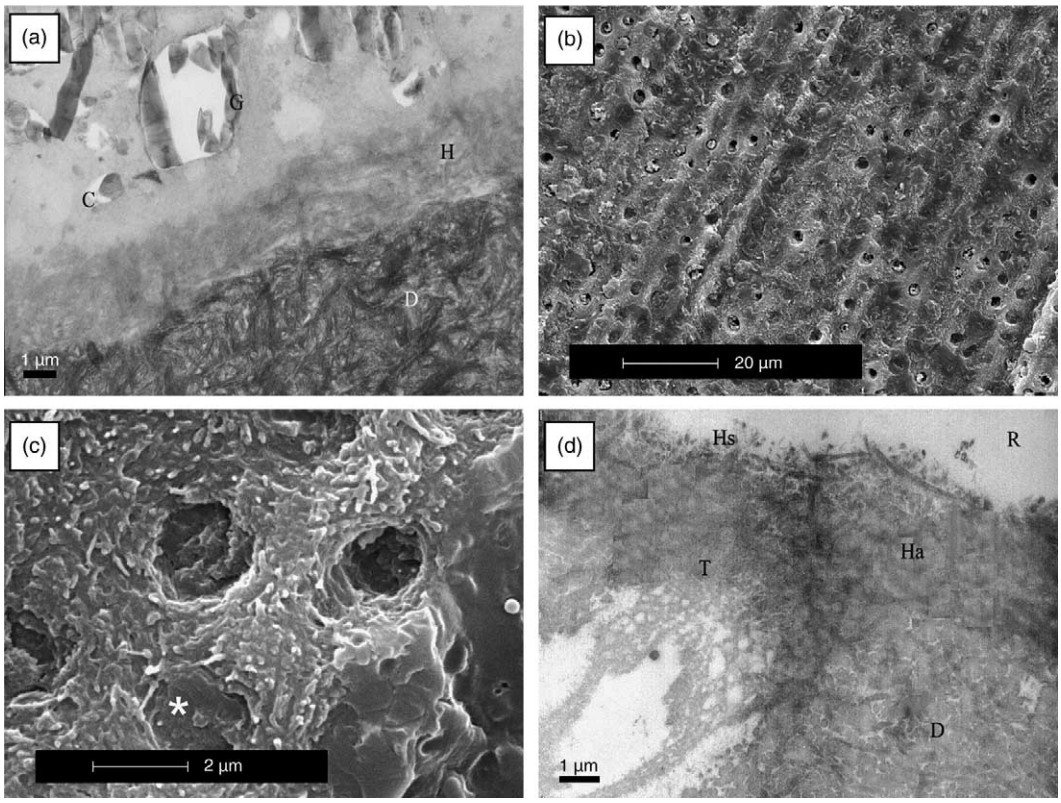

Figure 5 Interface and fractography analysis of group PF (Panavia F 2.0). (a) TEM photomicrograph at 4000 magnification illustrating an overview of the interface between PF luting resin and superficial dentin. The hybrid layer is approximately 1.5–2 μm thick. (b) SEM micrograph at 600 magnification of adhesive failure from the deep dentin surface. (c) SEM micrograph at 4500 magnification of the same specimen as in (b). The failure occurred at the top of hybrid layer with cohesively fractured resin tags occluding the tubules (asterisk). (d) TEM photomicrograph at 5000 magnification illustrating the failure within the hybridized smear layer and smear plug in deep dentin. C, luting resin; H, hybrid layer; Ha, authentic hybridized dentin; Hs, hybridized smear layer; T, Resin tag; G, glass filler particle; D, demineralized dentin in the preparation of TEM; R, embedding resin.

fillers) (Technical Product Profile. 3M ESPE AG, Germany) and highly viscous luting resin. The smear layer and underlying dentin have been regarded as solid buffers that probably rapidly buffer the acidity of viscous solutions, thereby limiting the etching ability of acidic monomers (David H. Pashley, personal communication). The inability of RU to penetrate demineralized dentin is supported by SEM observation of insufficient infiltration of resin into the collagen network (Fig. 6). Since the HL was very thin to nonexistent, the μTBS of RU to regional dentin sites was relatively low, even in superficial dentin (Fig. 6a).

Theoretically, in deep and cervical dentin the decreased amount of intertubular dentin available limits the contribution of the HL to the μTBS, while the increased number and diameter of the tubules increases the cross-sectional area and volume of the resin tags. Therefore, the cohesive strength of the resin tags and the hybridization of resin tags to tubular walls play an important role in determining the bond strength in deep dentin [22]. For specimens in the SB group, after the peritubular dentin was etched with 10–3 solution, the circumferentially oriented collagen fibrils that line the tubule walls were completely exposed (Fig. 3). This allowed adhesive resin to infiltrate into the adjacent intertubular dentin and form hybridized resin tags with many branches [22]. Partially oxidized tri-*N*-butyl-borane (TBB), as the polymerization initiator in Super-Bond C&B luting resin system, utilizes oxygen and water to initiate radical polymerization of the resin monomers [23]. Therefore, when using SB for bonding to deep water-rich dentin, polymerization should be enhanced at the interface of hydrated dentin with resin, and continue outward into the luting resin. This moisture tolerance and interfacial polymerization of SB results in thorough polymerization and improvement of regional bond strength near the pulp [24]. This proposition is supported in the current study by the high μTBS to deep and cervical

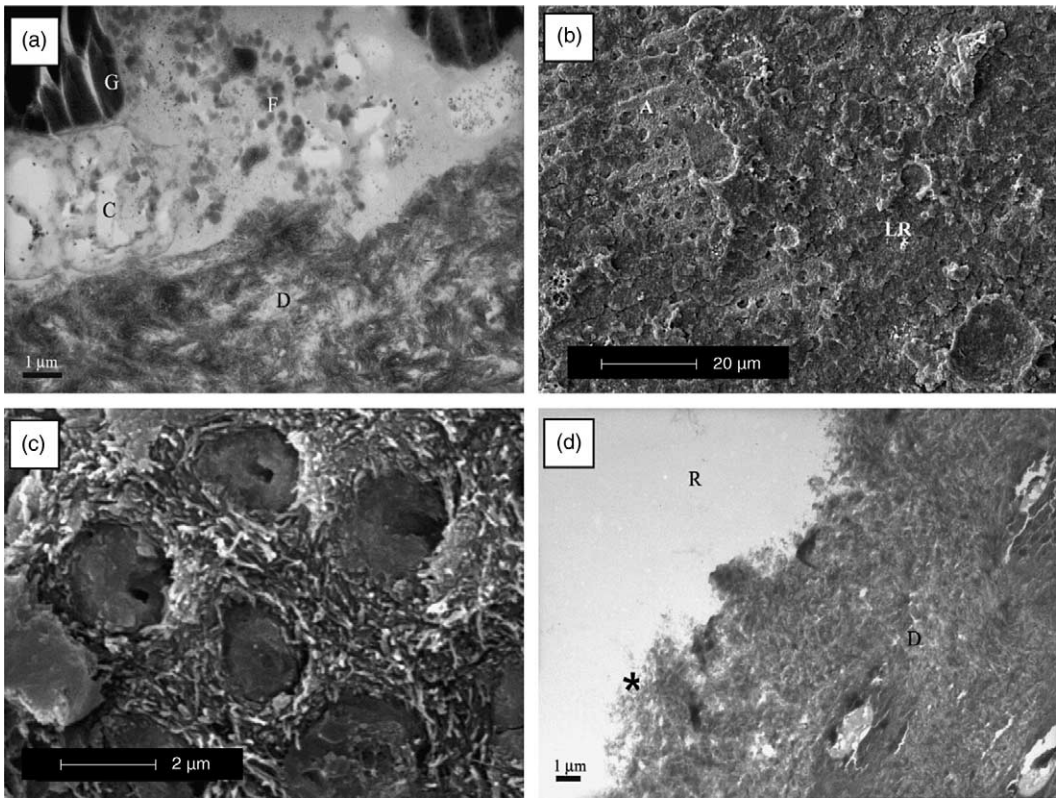

Figure 6 Interface and fractography analysis of a specimen in group RU (RelyX Unicem). (a) TEM photomicrograph at 5000 magnification illustrating an overview of the interface between RU luting resin and superficial dentin. The smear layer appears to be completely dissolved, but no obvious hybrid layer is observed. (b) SEM micrograph at 600 magnification of a debonded deep dentin specimen that adhesively failed at the top of dentin surface with a thin layer of luting resin (LR) remaining on the dentin surface. (c) SEM micrograph at 4500 magnification of adhesive failure at 'A' in the same specimen as in (b). The loose collagen fibrils in the intertubular dentin do not seem to be adequately enveloped by luting resin. (d) TEM photomicrograph at 3000 magnification illustrating the adhesive failure from the demineralized cervical dentin surface. The collagen fibrils along the fractured surface appear to be stretched into loose microfibrils (asterisk) without resin infiltration. C, luting resin; D, demineralized dentin in the preparation of TEM; G, glass filler particle; F, nanofiller; R, embedding resin.

dentin and TEM observation of the extension of hybridized resin tags into the tubules (Fig. 4). It might be concluded that well-hybridized resin tags contribute to the total micromechanical retention and bonding strength in dentin bonded with SB, especially in deep or cervical dentin [22].

However, this mechanism of adhesion might not be directly applied to self-etching systems. In the present study, the mean μTBS of PF to deep dentin and cervical dentin were significantly lower than that of superficial dentin. It was discovered that ED primer 2.0 did not completely remove smear plugs (Fig. 3). Therefore, PF luting resin probably penetrated into the residual smear plug to the partially demineralized collagen network around the tubular walls to form a thin bonding interface (Fig. 5d). However, this thin bonding of PF to the walls of tubules was strong enough to make the hybridized smear plugs and resin tags fracture at the tubule orifice during μTBS testing instead of being pulled out from the tubules (Fig. 5c). It can be concluded that the top of the hybridized smear layer became the weak link during μTBS testing supported the conclusions of others [25,26]. In the current study, since PF cured in its self-curing mode, water may have had time to diffuse from the tubules through the self-etching primed smear plugs to form water droplets at the bonding interface between dentin surface and luting resin during the curing time. These water droplets might function as sites of stress concentration when specimens were stressed to failure.

Water is an important ingredient for self-etching systems to ionize the acid and dissolve the minerals of the smear layers and dental hard tissues [27]. However, if any residual water is not sufficiently removed during the bonding procedures of self-etching systems, water would

compete with the monomers infiltrating into the demineralized zone to occupy the space on the demineralized collagen [28]. Such an 'overwet' condition might result in a dilution of the monomer concentration and interfere with the degree of polymerization of the resin [27,29]. This is perhaps one reason for the lower μTBS of the two self-etching systems to deep and cervical dentin than to superficial dentin.

Compared to groups SB and PF, a rather high percentage of partial adhesive failures that left a thin layer of cohesively fractured luting resin was found in all RU groups, indicating that the adhesion of luting resin to dentin was rather weak. A possible reason is that the self-curing polymerization of RU is not complete, although a prolonged storage time was used, which is supported by the low degree of conversion (26%) reported for self-cured RU [30]. Therefore, even if RU is a heavily filled composite resin, its strength is not high if it is not completely polymerized, which might be also one reason for the low μTBS in RU groups.

In cervical dentin, the resin tags penetrated into oblique tubules to provide non-parallel retention [9]. This might account for a differing μTBS as well as the increase in cohesive failures in the three luting resins in cervical dentin compared to deep dentin, although no statistical difference was detected in the present study. Therefore, the first null hypothesis that μTBSs of three luting resins to dentin do not vary with the dentin location has to be partially rejected. In addition, the variations in mechanical and structural properties in dentin from the DEJ to the pulp could influence the dentin bond strength [31].

In conclusion, our results proved that luting resins with different chemical formulations and application techniques yield morphologically different interfacial microstructures and regional dentin bond strengths. Therefore, the second null hypothesis that the different chemical formulations of the three luting resins and applications do not result in a different morphological appearance of the bonding interface has to be rejected.

Acknowledgements

This study was supported by Sun Medical Co. Ltd (Shiga, Japan). The authors greatly appreciate Prof. D.H. Pashley for critically reviewing the manuscript. We thank Mr Lehmann and Ms Marquardt for their technical guidance in the preparation and observation of TEM and SEM. We also thank Kuraray Medical Inc. (Osaka, Japan) and 3M ESPE AG (Seefeld, Germany) for their generous supply of materials.

References

[1] McCabe JF, Walls AWG. *Application of dental materials*. 8th ed. Malden: Blackwell Science; 1998. p. 189–201.
[2] Nakabayashi N, Pashley DH. *Hybridization of dental hard tissues*. Tokyo: Quintessence; 1998. p. 16–89.
[3] Pashley DH, Carvalho RM. Dentin permeability and dentin adhesion: review. *J Dent* 1997;**25**:355–72.
[4] Marshall GW, Marshall SJ, Kinney JH, Balooch M. The dentin substrate: structure and properties related to bonding. *J Dent* 1997;**25**:441–58.
[5] Takahashi A, Sato Y, Uno S, Pereira PNR, Sano H. Effects of mechanical properties of adhesive resins on bond strength to dentin. *Dent Mater* 2002;**18**:263–8.
[6] Shono Y, Ogawa T, Terashita M, Carvalho RM, Pashley EL, Pashley DH. Regional measurement of resin–dentin bonding as an array. *J Dent Res* 1999;**78**:699–705.
[7] Burrow MF, Takakura H, Nakajima M, Inai N, Tagami J, Takatsu T. The influence of age and depth of dentin on bonding. *Dent Mater* 1994;**10**:241–6.
[8] Mixson JM, Spencer P, Moore DL, Chappel RP, Adams S. Surface morphology and chemical characterization of abrasion/erosion lesions. *Am J Dent* 1995;**8**:5–9.
[9] Ferrari M, Cagidiaco MC, Vichi A, Mannocci F, Mason PN, Mjor IA. Bonding of all-porcelain crown: structural characteristics of the substrate. *Dent Mater* 2001;**17**:156–64.
[10] Swift EJ. Enamel–dentin bonding. *J Esthet Dent* 2003;**15**: 203.
[11] Bouillaguet S, Gysi P, Wataha JC, Ciucchi B, Cattani M, Godin C, et al. Bond strength of composite to dentin using conventional, one-step, and self-etching adhesive systems. *J Dent* 2001;**29**:55–61.
[12] Van Meerbeek B, Yoshida Y, Snauwaert J. Hybridization effectiveness of a two-step versus three step smear layer removing adhesive material examined correlatively by TEM and AFM. *J Adhes Dent* 1999;**1**:7–23.
[13] Sano H, Shono T, Sonoda H, Takatsu T, Ciucchi B, Carvalho R, et al. Relationship between surface area for adhesion and tensile bond strength-evaluation of a micro-tensile bond test. *Dent Mater* 1994;**10**:236–40.
[14] Pashley DH, Carvalho RM, Sano H, Nakajima M, Yoshiyama M, Shono Y, et al. The microtensile bond test: a review. *J Adhes Dent* 1999;**1**:299–309.
[15] Mak YF, Lai SCN, Cheung GSP, Chan AWK, Tay RT, Pashley DH. Micro-tensile bond testing of luting resins to dentin and an indirect resin composite. *Dent Mater* 2002; **18**:609–21.
[16] Watanabe I, Nakabayashi N, Pashley DH. Bonding to ground dentin using a self-etching Phenyl-P primer. *J Dent Res* 1994;**73**:1212–20.
[17] Koibuchi H, Yasuda N, Nakabayashi N. Bonding to dentin with a self-etching primer: the effect of smear layers. *Dent Mater* 2001;**17**:122–6.
[18] Feilzer AJ, De Gee AJ, Davidson CL. Setting stress in composite resin in relation to configuration of the restoration. *J Dent Res* 1987;**66**:1636–9.
[19] Bouillaguet S, Troesch S, Wataha JC, Krejci I, Meyer JM, Pashley DH. Microtensile bond strength between adhesive cements and root canal dentin. *Dent Mater* 2003;**19**: 199–205.

[20] Phrukkanon S, Burrow MF, Tyas MJ. The influence of cross-sectional shape and surface area on the microtensile bond test. *Dent Mater* 1998;**14**:212–21.
[21] Ohno H, Kimura M, Fuchigamis Oguri M. Dental composition. United States Patent No. 5,739,177, April 14; 1998.
[22] Pashley DH, Ciucchi B, Sano H, Carvalho RM, Russell CM. Bond strength versus dentin structure: a modelling approach. *Arch Oral Biol* 1995;**40**:1109–18.
[23] Okamoto Y, Takahata K, Saeki K. Studies of the behaviour of partially oxidized tributylborane as a radical initiator for methyl methacrylate (MMA) polymerization. *Chem Lett, The Chemical Society of Japan* 1998;1247–8.
[24] Ozturk AN, Belli S, Eskitascioglu G. The in vitro effect of pulpal pressure and luting agent on tensile bond strength of complete cast crowns. *J Prosthet Dent* 2004; **91**:253–7.
[25] Carvalho RM, Pegoraro TA, Tay FR, Pegoraro LF, Silva NR, Pashley DH. Adhesive permeability affects coupling of luting resins that utilise self-etching primers to dentin. *J Dent* 2004;**32**:55–65.
[26] Jayasooriya PR, Pereira PN, Nikaido T, Barrow MF, Tagami J. The effect of a resin coating on the interfacial adaptation of composite inlays. *Oper Dent* 2003;**28**:28–35.
[27] Jacobson T, Söderholm K-J. Some effects of water on dentin bonding. *Dent Mater* 1995;**11**:132–6.
[28] Armstrong SR, Keller JC, Boyer DB. Mode of failure in the dentin–adhesive resin–resin composite bonded joint as determined by strength-based (μTBS) and fracture-based (CNSB) mechanical testing. *Dent Mater* 2002;**17**:201–10.
[29] Pereira PNP, Okuda M, Sano H, Yoshikawa T, Burrow MF, Tagami J. Effect of intrinsic wetness and regional difference on dentin bond strength. *Dent Mater* 1999;**15**:46–53.
[30] Kumbuloglu O, Lassila LV, User A, Vallittu PK. A study of the physical and chemical properties of four resin composite luting cements. *Int J Prosthodont* 2004;**17**:357–63.
[31] Tesch W, Eidelman N, Roschger P, Goldenberg F, Klaushofer K, Fratzl P. Graded microstructure and mechanical properties of human crown dentin. *Calcif Tissue Int* 2001;**69**:147–57.