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Influence of different drying methods on microtensile bond strength of self-adhesive resin cements to dentin

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Abstract

Objective. This study investigated the effect of different drying methods of dentin surface on the bonding efficacy of self-adhesive resin cements (SRCs). ***Materials and methods.*** Three SRCs (RelyX U200, RU; Maxcem Elite, ME; and BisCem, BC) and one resin-modified glass ionomer cement (RelyX Luting 2, RL) were used. The characteristics of the materials were evaluated using thermogravimetric analysis and surface roughness and contact angle measurements. Human dentin surfaces were finished with 600-grit silicon carbide paper and assigned to three groups according to these drying methods: ethanol dehydration, drying by waiting for 10 s after blot-drying and blot-drying. The four cements were used for luting composite overlays to the dried dentin. After 24 h storage at 37°C and 100% relative humidity, stick-shaped specimens with a cross-sectional area of 0.8 mm^2 were prepared and stressed to failure in tension at a crosshead speed of 0.5 mm/min ($n = 27$). Failure modes of fractured specimens were assessed by optical and scanning electron microscopy. ***Results.*** RL was the most hydrophilic, followed by BC and ME and then RU. All the luting cements luted to ethanol-dehydrated dentin showed zero bond strengths. For the three SRCs, drying by waiting produced higher microtensile bond strengths than blot-drying. RU showed the best bonding performance in the above two dentin conditions. RL showed significantly higher bond strength in blot-drying condition than in drying-by-waiting ($p < 0.001$). ***Conclusions.*** This study suggests that dentin surface moisture has a crucial effect on the bond strength of SRCs.

Key Words: *dentin wetness, microtensile bond strength, self-adhesive resin cement, surface energy parameters*

Introduction

Resin cement development has been based on the chemistry of resin composites and adhesives [1]. Recently, self-adhesive resin cements (SRCs) have been introduced in an attempt to simplify the cementation procedure and, thus, to reduce application time and technique sensitivity [2,3]. They have recently won over dental clinicians due to benefits such as the simplification of clinical steps, low incidence of post-operative sensitivity and early clinical success [3,4].

Although the basic composition of SRCs is similar to conventional resin cement, they also contain additional acidic functional monomers to condition the tooth surface and to potentially induce chemical adhesion to tooth substrate [3,5]. Similar to that of glass ionomer cements, the setting reaction of SRCs involves the acid-base reaction within an aqueous environment [6]. For this reason, the manufacturer recommends avoiding overdrying the dentin surface when using these cements.

However, the effect of tooth surface moisture on the bond strength of SRCs remains controversial [5]. According to Hiraishi et al. [7], the microtensile bond strengths of a SRC (RelyX Unicem, 3M ESPE, Seefeld, Germany) to dentin were low and not significantly different between the absence/presence of pulpal pressure. Guarda et al. [8] reported higher bond strength of a SRC (RelyX Unicem) on moist dentin than dry dentin, suggesting removing only excess dentin moisture rather than air drying.

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Although most manufacturers of SRCs suggest that the tooth surface be lightly or strongly dried with compressed air (Table I), such instructions often seem too vague for clinicians to apply.

It has been shown that SRCs do not sufficiently etch through clinically relevant smear layers into the underlying intact dentin and, thus, interact only superficially with dentin [9,10], notwithstanding the lower initial pH than conventional resin cements [11]. Due to the limited micromechanical retention, the bonding of SRC to tooth substrate may be more dependent on a chemical interaction between the acidic monomers and the calcium in the hydroxyapatite [3]. Accordingly, we assumed that the bonding performance of SRCs to dentin may be determined primarily by the surface chemical interaction of dentin with SRCs. Although dental restorations do not always require adhesive bonding to tooth structure, strong adhesion is still desirable in many clinical situations, especially when the prepared tooth structure is unusually short or tapered [12].

This study examined the microtensile bond strengths of three SRCs and one resin-modified glass ionomer cement (RMGIC) (for comparison) to dentin subjected to three different drying procedures. The luting cements were characterized by surface roughness and contact angle (CA) measurements. Relative differences in water amount on the dentin surfaces were evaluated by Fourier transform infrared (FTIR) spectroscopy. The null hypothesis tested was that different drying methods would not influence the microtensile bond strength of the cements to dentin.

Materials and methods

Luting cements

Three commercially available SRCs (RelyX U200, RU; Maxcem Elite, ME; and BisCem, BC) and one RMGIC (RelyX Luting 2, RL) were investigated. Their manufacturers, compositions and original

Table I. Three self-adhesive resin cements and one resin-modified glass ionomer cement tested.

Brand name (code, manufacturer)	Composition* (batch number)	Filler loading/average particle size*	Original instructions for preparation of the tooth surface prior to cementation*
RelyX U200 (RU, 3M ESPE, Seefeld, Germany)	Base: mixture of mono-, di- and tri-glycerol esters of phosphoric acid dimethacrylate, TEGDMA, silane-treated glass, silane-treated silica, glass, sodium persulfate, tert-butyl peroxy-3,5,5-trimethylhexanoate; Catalyst: substituted dimethacrylate, 1,12-dodecane dimethacrylate, silane-treated glass, silane-treated silica, calcium hydroxide, calcium salt of 1-benzyl-5-phenyl-barbic-acid, sodium p-toluenesulfinate (492696)	43 vol%/12.5 μm	Clean and rinse. Lightly air dry in 2–3 bursts of air or use cotton pellets to dry it off. The surface has a slightly glossy appearance
Maxcem Elite (ME, Kerr Corp., Orange, CA)	GPDM, TEGDMA, fillers, ytterbium fluoride, activators, stabilizers, HEMA, cumene hydroperoxide, titanium dioxide, pigments (4678975)	69 wt%	Clean and rinse. Air dry (do not desiccate)
BisCem (BC, Bisco Inc., Schaumberg, IL)	Di-HEMA phosphate, Tetra-EGDMA, glass (1300000248)	Base: 36 vol (60 wt)%; Catalyst: 40 vol (62 wt)%/Base: 1.0 μm; Catalyst 3.5 μm	Clean and rinse. Remove all surface water using a strong stream of air on the preparation for 3–5 s. Do not desiccate
RelyX Luting 2 (RL, 3M ESPE, St. Paul, MN)	Paste A: fluoroaluminosilicate glass, proprietary reducing agent, HEMA, water; Paste B: methacrylated polycarboxylic acid, Bis-GMA, HEMA, water, potassium persulfate, zirconia silica filler (N316506)		Clean and rinse. Dry with air, cotton or a paper tip. Leave tooth surface moist. Do not overdry

*Manufacturer supplied.

TEGDMA, triethylene glycol dimethacrylate; GPDM, glycerol phosphate dimethacrylate; HEMA, 2-hydroxyethyl methacrylate; Di-HEMA phosphate, di-2-hydroxyethyl methacryl hydrogenphosphate; Tetra-EGDMA, tetraethylene glycol dimethacrylate; Bis-GMA, bisphenol A diglycidyl methacrylate.

instructions for the tooth surface preparation prior to the cementation are summarized in Table I.

Thermal analysis

Each mixed luting cement was positioned in a platinum sample holder. The sample holder containing the material was covered with a polyester strip and glass slide. RL was allowed to self-cure; the three SRCs were additionally irradiated for 40 s by placing the tip of the light guide of a quartz-tungsten-halogen curing light (Elipar TriLight, 3M ESPE; standard mode, output intensity = 750 mW/cm^2) against the glass slide. Thermal gravimetric analysis was initiated at room temperature and increased to 800°C with a heat rate of 10°C/min at 50 mL/min under nitrogen flow [13]. The weight (%) of the samples in the range of 25–800°C was recorded.

Surface roughness and surface energy parameters

Cylindrical molds (10 mm in diameter; 1 mm in height) were placed on a polyester strip over a glass slide. Each mixed cement was filled into the mold, then covered with another polyester strip and glass slide. RL was allowed to self-cure; all the three SRCs were additionally irradiated for 40 s by placing the tip of the light guide of the curing light (Elipar TriLight) against the upper glass slide. After 10 min, the strips were removed from the specimen and the average surface roughness, R_a, of each specimen was measured using a profilometer (Surftest SV-400, Mitutoyo Corp., Kawasaki, Japan) at a stylus speed of 0.1 mm/s, a cut-off of 0.8 mm and a range of 600 μm. Five readings were performed for each specimen and the average was calculated as the R_a value of the specimen (five specimens for each cement).

Surface energy characteristics can be calculated by measuring the CAs of a liquid triplet with known surface energy parameters on solid surfaces [3,14]. Surface energy components of solid are calculated using an equation which combines the Young-Dupré equation with the Lifshitz-van der Waals/Lewis acid-base (LWAB) theory [3,14]: $\gamma_l(1 + \cos\Theta) = 2 [(\gamma_s^{LW}\gamma_l^{LW})^{1/2} + (\gamma_s^{+}\gamma_l^{-})^{1/2} + (\gamma_s^{-}\gamma_l^{+})^{1/2}]$, where the superscripts LW, + and – refer to the Lifshitz-van der Waals, acid and base components, respectively. The total surface energy γ_s is derived by the equation [3,14]: $\gamma_s = \gamma_s^{LW} + \gamma_s^{AB} = \gamma_s^{LW} + 2(\gamma_s^{+}\gamma_s^{-})^{1/2}$, in which γ_s^{AB} is the acid/base component. The work of cohesion, W_c, can be expressed in terms of the free energy, G, so that $\Delta G_c = -2\gamma = -W_c$ [15]. The degree of hydrophilicity/hydrophobicity of a material is linked to the magnitude of $\Delta G_{sws} = -2\gamma_{sw}$, where $\gamma_{sw} = \gamma_{sw}^{LW} + \gamma_{sw}^{AB}$. The LW interfacial tension and the AB interfacial tension are calculated using the equations, respectively [3,16]: $\gamma_{sw}^{LW} = [(\gamma_s^{LW})^{1/2} - (\gamma_w^{LW})^{1/2}]^2$ and $\gamma_{sw}^{AB} = 2 [(\gamma_s^{+}\gamma_s^{-})^{1/2} + (\gamma_w^{+}\gamma_w^{-})^{1/2} - (\gamma_s^{+}\gamma_w^{-})^{1/2} - (\gamma_w^{+}\gamma_s^{-})^{1/2}]$. Five cured specimens per material were prepared in the same way as for the surface roughness measurement. The CA of each of three different liquids (water, glycerol and methylene iodide) was determined on the surfaces by the sessile drop method using a CA goniometer (OCA 15 plus, DataPhysics Instrument GmbH, Filderstadt, Germany) [17]. All CA measurements were performed in a temperature controlled room at 23 ± 1°C with relative humidity at 50 ± 5% [18].

Tooth preparation

Fifty-one non-carious human molars were collected after obtaining the patients' informed consents obtained under a protocol approved by the Ethics Committee of the School of Dentistry, Kyungpook National University. The teeth were stored in 0.5% chloramine in water at 4°C and used within 1 month following extraction. The occlusal enamel and roots of the teeth were removed using a slow-speed saw with a diamond-impregnated disk (Isomet, Buehler Ltd., Lake Bluff, IL) under water cooling to form 5–6 mm thick, parallel-sided crown segments [19]. A 600-grit silicon carbide (SiC) paper was used under running water to create a thin smear layer on the dentin surface [20].

The dentin substrates to be bonded were divided into three groups depending upon the drying methods. In the ethanol dehydration group, the teeth, after removing the contents of the pulp chamber, were dehydrated through a series of ascending ethanol concentrations (70%, 80%, 95%, three changes in 100%) for 2 h each and then left to completely dehydrate in absolute ethanol for an additional 48 h [19]. In the drying-by-waiting group, the specimens were blot-dried and then left (for 10 s) until all visible moisture was evaporated from the dentin surface under a stereomicroscope (SZ61, Olympus, Tokyo, Japan) at a magnification of 20× [19]. In the blot-drying group, the dentin surfaces were blot-dried and left inverted on a piece of wet, lint-free paper (Kimwipes, Kimberly-Clark Corp., Roswell, GA) until ready for luting with the cements [21].

FTIR spectroscopy

The relative amount of water remaining on the dried dentin surfaces was estimated using a FTIR spectrophotometer (IRPrestige-21, Shimadzu Corp., Kyoto, Japan) with an attenuated total reflectance (ATR) unit (MIRacle, Pike Technologies Inc., Madison, WI). Fifteen dentin specimens were used. Immediately after being subjected to the drying procedures described above, the dentin surfaces were put onto the face of the ATR surface. The absorbance spectra were acquired by scanning the specimens 10 times over a 4000–750 cm^{-1} range at a resolution of 4 cm^{-1}. After atmospheric and baseline corrections, the net peak absorbance height ratio of water (the hydrogen bonded O-H stretching vibration at 3396 cm^{-1}) to apatite (the phosphate ν_3 mode (ν_3PO_4) at 1026 cm^{-1}) was used to assess the relative amount of water [22,23].

Composite overlay preparation

Composite cylinders (10 mm in diameter and 4 mm in height) were prepared using a split aluminum mold by layering two 2-mm thick increments of a composite (Aelite All-Purpose Body, Bisco Inc., Schaumberg, IL; A2, lot #: 1300001789). Each increment was light-cured for 40 s with the curing light (Elipar Tri-Light). The bonding surface of each composite was ground with 600-grit SiC paper to create a roughened surface [7], etched with 32% phosphoric acid gel (Uni-Etch, Bisco Inc.), rinsed with water and dried with oil-free air.

Luting procedures

Thirty-six dentin specimens were used. Immediately after drying, the surface was covered with adhesive masking tape 50 µm thick with a hole (7 mm in diameter) cut. Thereafter, the luting cements manipulated according to the manufacturers' directions were applied to the bonding surface of the composite discs and seated on the dentin surface using finger pressure. Cement excess was removed and SRCs were then additionally light-cured for 40 s from each side of the specimens to ensure optimal polymerization [20]. Luting procedures were performed at room temperature (23 ± 1°C) and relative humidity (50 ± 5%) [20]. All bonded specimens were then stored in a laboratory incubator for 24 h at 37°C and 100% relative humidity before sectioning [20,21].

Microtensile bond strength testing and examination of the fractured surfaces

Each bonded specimen was longitudinally sectioned into 0.9 mm-thick slabs with the slow-speed diamond saw (Isomet). Each slab was fixed on a glass platform with sticky wax and serially sectioned into 0.8 mm^2 (±0.1) sticks, in accordance with the 'non-trimming' method of the microtensile test [2,20]. The exact dimensions of each stick were measured using a digital caliper to calculate the precise cross-sectional area. For microtensile bond strength testing, nine sticks were sectioned from the center of each bonded specimen [7]. Thus, the use of three teeth resulted in a total of 27 sticks for each of the groups tested. Sticks with premature bond failure were assigned a null bond strength value and were included in the compilation of the mean bond strength as well as the failure mode assessment [21]. The bonded composite-dentin sticks were attached to a testing device with cyanoacrylate glue (Zapit, DVA, Corona, CA). The device was attached to a microtensile bond tester (Bisco Inc.) and loaded in tension at a crosshead speed of 0.5 mm/min until failure [21].

After fracturing, all specimens were examined under the stereomicroscope (SZ61) at a magnification of 20× and failure modes were categorized as: (a) adhesive failure along the dentin–cement interface; (b) adhesive failure along the cement–composite interface; (c) mixed failure involving cohesive failure of the cement and adhesive failure along a or b; (d) cohesive failure of the dentin; and (e) cohesive failure of the composite [24]. Representative fractured sticks (dentin sides) were additionally examined under a field emission-scanning electron microscope (FE-SEM, JSM-6700F, Jeol, Tokyo, Japan) after air-drying and sputter-coating with platinum [7].

Statistical analysis

All the data were statistically analyzed by non-parametric methods at $\alpha = 0.05$ because they did not meet the homogeneity of variances assumption (Levene's test). The Kruskal-Wallis test was used to compare the groups, followed by the Mann-Whitney post-hoc test, with adjustment of significance levels using the Sidak correction for multiple testing [25]. All the statistical analyses were performed using SPSS 17.0 for Windows (SPSS Inc, Chicago, IL).

Results

Thermogravimetric analysis

The thermogravimetric behavior of the luting cements tested is presented in Figure 1. Although the weight loss trends of the three SRCs were similar, ME

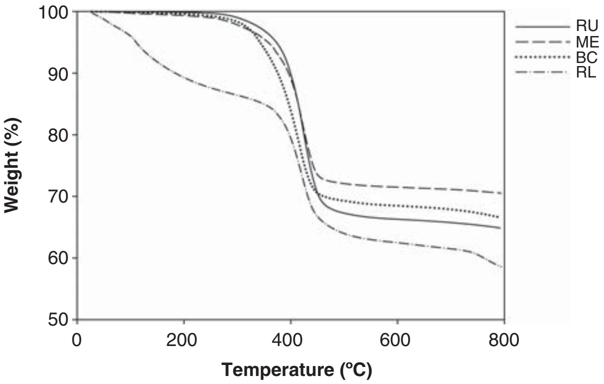

Figure 1. Representative curves of the thermogravimetric analysis of the four luting cements tested in this study. RU, RelyX U200; ME, Maxcem Elite; BC, BisCem; RL, RelyX Luting 2.

Table II. Mean (SD) of surface roughness and surface energy parameters of the luting cements tested (n = 5).

	Surface roughness (μm)	Surface energy parameters (mJ/m^2)					
Luting cement	R_a	γ_s	γ_s^{LW}	γ_s^{+}	γ_s^{-}	γ_s^{AB}	ΔG_{sws}
RU (RelyX U200)	0.10 (0.01)^{a}	51.12 (0.50)^{a}	46.68 (0.53)^{a}	0.29 (0.14)^{a}	18.20 (1.48)^{a}	4.44 (0.99)^{a}	−23.54 (2.55)^{a}
ME (Maxcem Elite)	0.11 (0.01)^{a}	48.14 (1.87)^{a}	41.20 (1.27)^{b}	0.25 (0.06)^{a}	48.31 (1.42)^{b}	6.94 (0.89)^{b}	28.45 (1.12)^{b}
BC (BisCem)	0.08 (0.02)^{a}	51.70 (0.96)^{a}	44.57 (1.14)^{b}	0.25 (0.04)^{a}	50.07 (1.01)^{b}	7.12 (0.57)^{b}	28.78 (0.91)^{b}
RL (RelyX Luting 2)	0.21 (0.03)^{b}	84.41 (4.16)^{b}	43.73 (1.33)^{b}	4.68 (0.94)^{b}	88.89 (3.16)^{c}	40.68 (4.70)^{c}	43.01 (1.85)^{c}

Within a column, values with the same superscript letter indicate no statistically significant difference according to Kruskal-Wallis and Mann-Whitney tests with the Sidak correction at $\alpha = 0.05$.

γ_s, total surface energy; γ_s^{LW}, Lifshitz-van der Waals component; γ_s^{+}, acid component; γ_s^{-}, base component; γ_s^{AB}, acid/base component; ΔG_{sws}, degree of hydrophobicity/hydrophilicity.

showed a more rapid loss at early stage (in the temperature range of 25–300°C), followed by BC and then RU. RL showed a considerably different weight loss pattern from the SRCs.

Surface characteristics of the cements

Table II summarizes the surface roughness and surface energy parameters of the luting cements as calculated from the CA data. RL showed a significantly higher R_a value ($p = 0.047$) and significantly greater surface energy parameter values ($p < 0.05$) than the three SRCs except for γ_s^{LW}. RU exhibited smaller γ_s^{-}, γ_s^{AB}, and ΔG_{sws} values than the other two SRCs ($p < 0.05$), whose surface energy parameter values were statistically similar to each other ($p > 0.05$).

FTIR analysis of the dentin surfaces

FTIR spectroscopic results showing the relative amount of water remaining on the dried dentin surface are represented in Figure 2 and Table III. In the ethanol dehydration group, little O-H peak was detected. According to the water/apatite ratio, blot-dried dentin surface was found to contain ~3-times of water when compared to dentin dried by waiting.

Microtensile bond strength

Table IV summarizes microtensile bond strengths of the four luting cements to the dried dentin. The ethanol dehydration group consistently resulted in zero bond strengths, regardless of the luting cements

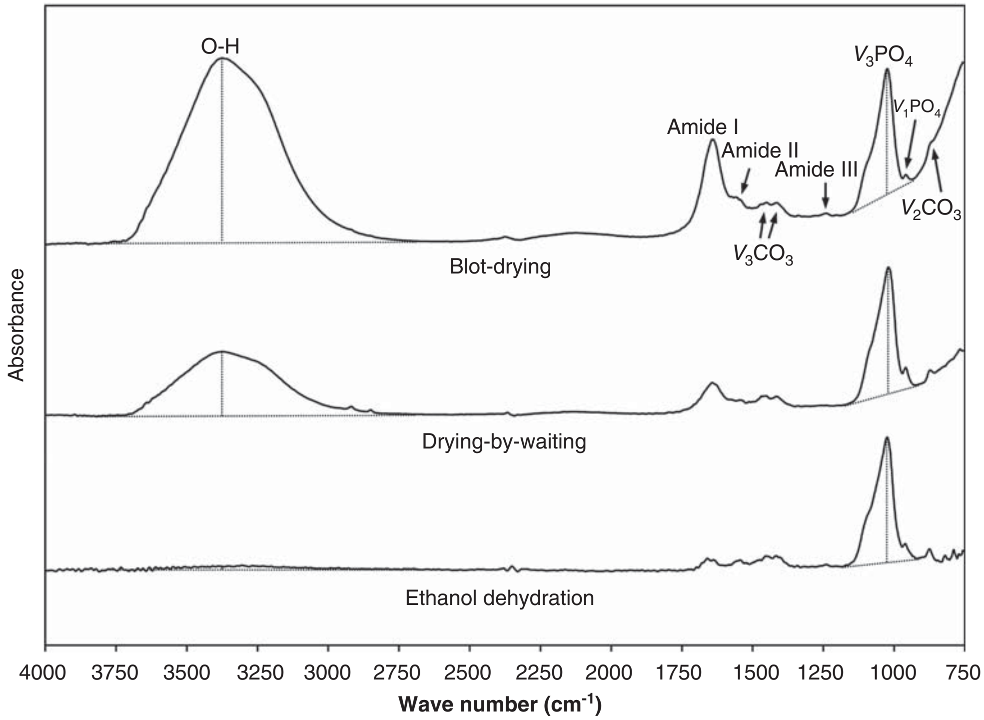

Figure 2. Representative FTIR absorbance spectra of dentin surface subjected to three different drying methods. The relative amount of the surface water was calculated using the peak height ratio of O-H (3396 cm^{-1}) to ν_3PO_4 (1026 cm^{-1}).

Table III. Mean (SD) of relative amounts of the dentin surface water calculated from the FTIR spectra (n = 5).

Drying methods	Peak height ratio of O-H/ν_3PO$_4$
Ethanol dehydration	0.03 (0.01)^a
Drying-by-waiting	0.51 (0.16)^b
Blot-drying	1.47 (0.40)^c

Values with the same superscript letter are not significantly different according to Kruskal-Wallis and Mann-Whitney tests with the Sidak correction at $\alpha = 0.05$.

used. In the drying-by-waiting group, RU showed the highest value (mean = 29.9 MPa) among all the SRCs tested, the difference being statistically significant ($p < 0.001$), whereas RL produced a significantly lower value (mean = 1.1 MPa) than the others ($p < 0.001$). The bond strengths significantly decreased when the SRCs were bonded to blot-dried dentin ($p < 0.001$), although RU still produced the highest value (mean = 8.0 MPa). In contrast, the bond strength of RL was significantly higher in the blot-drying group than in the drying-by-waiting group ($p < 0.001$).

Failure analysis

Figure 3 shows the failure pattern distribution (%) as assigned using the light microscope. The ethanol dehydration group produced only pre-testing failures. In the drying-by-waiting group, the failure mode for SRCs was mostly mixed or adhesive along the dentin–cement interface, whereas pre-testing failure was predominant for RL. In the blot-drying group, the incidence of adhesive failure increased for RU, while pre-testing failure was predominant for the other two SRCs. Failure modes for RL luted to blot-dried dentin were either adhesive failure along the cement–composite interface or mixed.

Figure 4 shows the representative SEM micrographs of the fractured dentin sides. In a specimen from the RU/drying-by-waiting group, the cement-covered surface (cohesive failure of the cement) showed the formation of numerous voids (Figure 4A). In a specimen from the ME/drying-by-waiting group, an adhesive failure region along the dentin–cement interface revealed mild demineralization of the smear layer (Figure 4B). Figures 4C and D show mixed failure involving adhesive failure along the dentin–cement interface and along the cement–composite interface in specimens from BC and RL luted to blot-dried dentin, respectively.

Discussion

This *in-vitro* study showed that the dentin surface moisture remaining after drying crucially affected the bond strength of the SRCs as well as the RMGIC (Table IV, Figure 3). Thus, the null hypothesis that different drying methods would not influence the microtensile bond strength of the cements to dentin was rejected. The three SRCs showed the best bonding performance when only a small amount of water remained on the dentin surface (Table IV). This condition was achieved by waiting 10 s for water to evaporate from the dentin surface.

This study used FTIR spectroscopy to estimate the relative amount of water on the dentin surface subjected to different drying methods (Figure 2 and Table III). The spectrum of ethanol dehydration clearly showed that the drying procedure almost completely removed water from the dentin specimen.

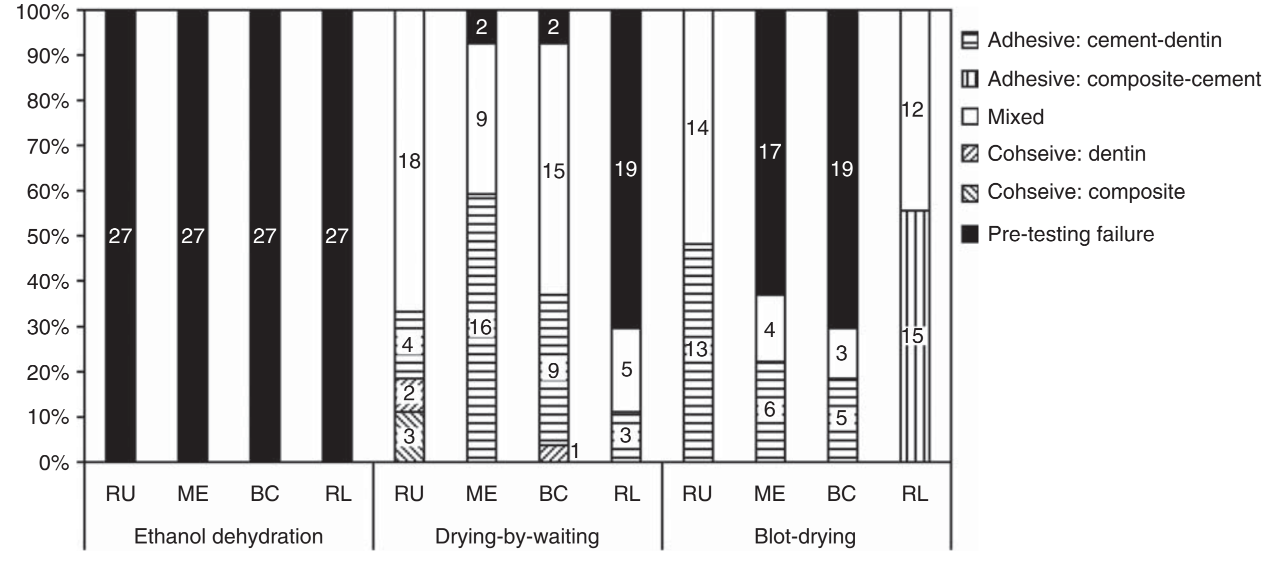

Figure 3. Type of failure mode as identified with a light microscope at 20× magnification and calculated as a percentage. Numbers inside the bars represent the incidence (number of sticks) of each type of failure per group (n = 27). RU, RelyX U200; ME, Maxcem Elite; BC, BisCem; RL, RelyX Luting 2.

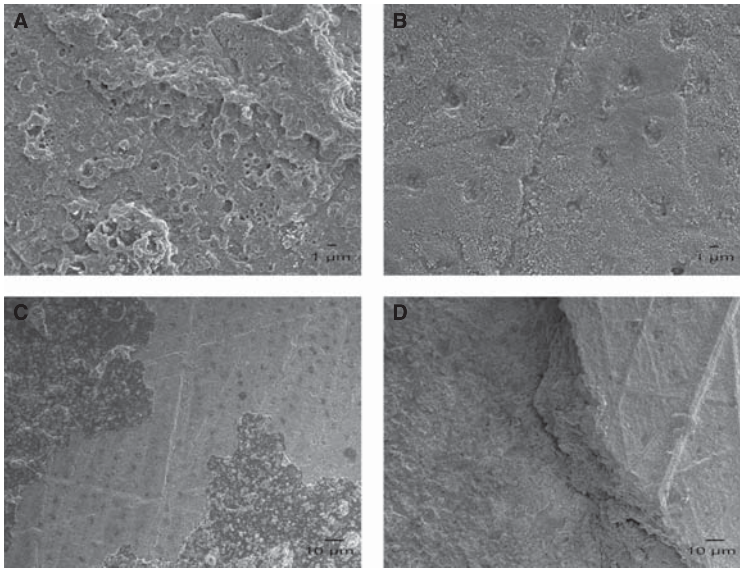

Figure 4. Representative SEM micrographs of fractured dentin sides. (A) RelyX U200 luted to dried dentin by waiting (RU/drying-by-waiting), showing cement-covered surface with formation of numerous voids (3000×). (B) Maxcem Elite luted to dried dentin by waiting (ME/drying-by-waiting), showing mild demineralization of smear layer (3000×). (C) BisCem luted to blot-dried dentin (BC/blot-drying), showing mixed failure involving cohesive failure of the cement and adhesive failure along the dentin–cement interface (650×). (D) RelyX Luting 2 luted to blot-dried dentin (RL/blot-drying), showing mixed failure involving cohesive failure of the cement and adhesive failure along the cement–composite interface (650×).

Although this dentin condition is not clinically relevant, the zero bond strengths and 100% pre-testing failures for the SRCs confirm that a certain amount of water should exist on the dentin surface for bonding with the cements (Table IV and Figure 3). Although the exact constituents of the SRCs tested are not fully disclosed by the manufacturers, SRCs generally contain no or only a little water in their chemical composition and water may only derive from the interaction of acid groups and alkaline filler or tooth apatite [20]. The thermogravimetric curves of the SRCs (Figure 1) suggest the materials contain little water as little weight loss was observed at early stage (up to 100°C) [26]. Acidic monomers of SRCs need water to become ionized and acid-etch and interact with dentin [20], ionization being more efficiently facilitated on hydrated dentin substrates [6]. It, thus, seems that the presence of a certain amount of water on the dentin surface is a pre-requisite for optimal bonding with SRCs.

In a pilot FTIR test, air-drying of the dentin surface generally showed less surface water than did blot-drying, but the results were less consistent than the dry-by-waiting method. On dentin dried by waiting (for 10 s) after blot drying, only a small amount of water remained on the surface (Figure 2 and Table III), which looked matte macroscopically. In this dentin surface condition, the highest bond strengths were obtained for all the SRCs (mean = 6.1–29.9 MPa) (Table IV). A strong bond is achieved only when functional groups in the acidic monomers of SRCs produce an optimal interaction with hydroxyapatite on the dentin surface [3]. Thus, the findings of this study suggest that only a little water suffices for the interaction of acid groups and tooth apatite, regardless of the absence/presence of water in SRCs.

The concentration of the acidic monomers in SRCs should be kept low enough to avoid excessive hydrophilicity in the final polymer, but high enough to

Table IV. Mean (SD) of microtensile bond strengths of the luting cements to dried dentin ($n = 27$).

	Drying methods of dentin		
Luting cement	Ethanol dehydration	Drying-by-waiting	Blot-drying
RU (RelyX U200)	0.0^{Aa}	$29.9\ (4.9)^{Ab}$	$8.0\ (2.5)^{Ac}$
ME (Maxcem Elite)	0.0^{Aa}	$6.1\ (3.2)^{Bb}$	$1.7\ (2.3)^{Bc}$
BC (BisCem)	0.0^{Aa}	$10.5\ (4.6)^{Cb}$	$1.4\ (2.4)^{Bc}$
RL (RelyX Luting 2)	0.0^{Aa}	$1.1\ (1.9)^{Db}$	$6.3\ (1.6)^{Cc}$

Values with the same superscript (capital letter: column; lowercase letter: row) are not significantly different according to Kruskal-Wallis and Mann-Whitney tests with the Sidak correction at $\alpha = 0.05$. The high standard deviations are due to inclusion of zero bond strengths for bonds which failed during handling prior to testing.

ensure an acceptable degree of self-etching and bonding to tooth [6]. In this study, the surface energy characteristics of the luting cements were investigated using CA measurements. Although uncured cements are applied to bonding substrates clinically, the surface energy parameters can be calculated from the CAs measured on cured luting cements. In such cases, it is assumed that surface energy parameters of monomer and polymer are the same or close because all groups and segments of the functional monomer molecules are also present in the polymeric material [3,14,27]. Since surface roughness may alter the contact angle values [28], surface roughness measurements were performed together. Although RL showed a significantly higher R_a than the three SRCs, there were only small variations (~0.1 μm) in the value (Table II). Surface roughness was, thus, assumed to have minimal influence on CAs [3].

The boundary between hydrophobicity and hydrophilicity of a solid material (s) in the presence of water (w) is equal to the cohesive polar attraction between the water molecules [3,16]. Taking $\Delta G_{sws} = 0$ as the boundary between hydrophobicity and hydrophilicity and classifying the luting cements according to the criterion [16], RU can be considered hydrophobic, whereas ME and BC are hydrophilic (Table II) [3]. The surface energy parameters of the cements and the microtensile bond strength values (Table IV) show that the bonding behavior of the cements depended not only on the wetness of dentin surface but also on the surface energy characteristics of the cements used. A more hydrophobic SRC (RU) showed better bonding performance to dried dentin by waiting than more hydrophilic ones (ME and BC) (Table IV). This likely indicates that a more than optimal amount of hydrophilic components may deteriorate bond strength by encouraging water absorption [3]. A significantly higher bond strength for BC than ME (mean = 10.5 vs 6.1 MPa, Table IV) implies that other factors besides surface energy characteristics (e.g. viscosity, type of acidic monomer, etc.) also affect the final bond strengths (Table IV) [10], an aspect requiring further investigation.

Blot-drying of dentin produced a significantly higher water/apatite ratio than drying-by-waiting, ~3:1 (Table III). The bond strengths of all the SRCs decreased significantly when the materials were luted to blot-dried dentin (Table IV), probably due to the presence of excess water at the interface [3]. Park [5] suggested that leaving a thin visible layer of water on the dentin surface is better than over-drying the dentin surface prior to use of SRCs. However, this dentin condition seems to contain too much water for optimal bonding with SRCs. Although the matrix resins in SRCs are more hydrophilic than in dental composite restoratives [6], the materials are basically hydrophobic. Therefore, too much water remaining on the dentin surface may unfavorably dilute the concentration of the acidic monomers and hinder the infiltration of the hydrophobic component of SRCs [5,7]. In the case of blot-dried dentin, the more hydrophobic RU still produced a significantly higher bond strength than the other two SRCs.

The bond strength results for RL indicate that an optimal amount of water on the dentin surface is also important for the bonding of RMGICs (Table IV). RL first showed weight loss at 25°C in the thermogravimetric curve (Figure 1), probably due to the large amount of water in its chemical composition. Moreover, its surface energy characteristics indicate the material is substantially hydrophilic and should be classified differently from the SRCs (Table II) [3,9,14]. Thus, the poor bonding for RL luted to dentin subjected to ethanol dehydration or drying-by-waiting may be due to poor wetting between the substantially hydrophilic cement and the over-dried hydrophobic dentin surfaces [6]. This study also suggests that blot-drying is more appropriate for RMGICs rather than SRCs. Although the RMGIC RL exhibited a bond strength statistically equivalent to that of RU, the failure mode was either mixed or adhesive along the cement-composite interface (Figures 3 and 4D), indicating favorable bonding between the dentin and the cement. Without proper moisture control of dentin prior to bonding, moisture-tolerant RMGIC would be preferable [29].

The present *in-vitro* study focused on surface interactions between luting cements and dentin with different wetness. It should be noted that the dentin surface and the cement curing conditions were not exactly the same as in clinical situations. The temperature and relative humidity in the oral cavity are higher than those in this laboratory study [30]. Such intra-oral conditions may influence not only the curing speed of the materials, but also dentin surface wetness and, consequently, may change the bond strength values. Moreover, the pulpal pressure was not simulated in this study. In vital dentin, an outward fluid flow from dentinal tubules under a slight positive pulpal pressure may hinder optimal seal at the bonded interfaces [20]. Nonetheless, the presence of numerous voids, probably filled with water, on the fractured dentin surface (Figure 4A) suggests that osmotic pressure still stimulated outward movement of tubular fluid [19]. When SRCs are left to cure in the self-cure mode, slower polymerization and lower final degree of conversion are more pronounced [31]. Thus, this phenomenon may have been more prominent when the SRCs were self-cured alone, possibly resulting in reduction in bond strength [20]. Although the results of this *in-vitro* study still cannot fully predict the clinical behavior of these materials applied on vital teeth, this study suggests that only a little dentin surface water is enough for SRCs to induce ionization of the acidic monomers followed by acid-base neutralization reactions involving the tooth and the basic filler [6].

Within the limitations of the present *in-vitro* study, it can be concluded that SRCs produce an acceptable bond strength only when the moisture of the dentin surface to be bonded is carefully controlled and dried. Although the optimal amount of moisture remaining on dentin surface was achieved by drying by waiting for 10 s after blot-drying, clinical trials are still needed to establish an optimal method of applying SRCs to tooth because intra-oral conditions are far more complex environments than those in *in-vitro* tests.

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