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Effect of self-curing activators and curing protocols on adhesive properties of universal adhesives bonded to dual-cured composites

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ABSTRACT

Objectives. To measure microshear bond strength (μSBS) and nanoleakage (NL) of self-etch universal adhesives under core buildup restorations using different curing protocols, at 24 h and after 6-month water storage.

Methods. Middle dentin of 55 molars was divided into: Clearfil Universal Bond [CFU], Prime&Bond Elect [PBE], and One Coat 7 Universal [OCU]. All-Bond Universal [ABU] and Clearfil SE Bond [CSE] were used as control. CFU, PBE and OCU were: light-cured [LC], dual-cured [DC] and self-cured [SC]. Data were analyzed separately (two-way ANOVA), Tukey's test ($\alpha = 0.05$).

Results. μSBS: At 24 h OCU/LC resulted in statistically higher μSBS than ABU. CSE/DC showed statistically higher μSBS than all DC adhesives. PBE/LC resulted in significant lower μSBS than the respective DC/SC modes ($p < 0.001$). At 6-month, both CFU and PBE (LC/SC), resulted in a significant decrease in μSBS. μSBS for OCU/DC decreased significantly ($p < 0.001$) compared to the respective LC/SC modes. NL: At 24 h, ABU showed %NL similar to CBU/LC and OCU/LC ($p > 0.05$). CSE/DC resulted in significantly higher %NL than OCU/DC but significantly lower than PBE/DC. CFU/LC/SC resulted in significantly lower %NL than CFU/DC. PBE/SC resulted in significant lower %NL than PBE/LC and PBE/DC. OCU/LC and OCU/DC resulted in significant lower %NL than OCU/SC ($p < 0.001$). At 6-month ABU, CSE, CFU/LC and CFU/SC, resulted in a significant increase in %NL.

Significance. Self-cured activator and different curing protocols influenced μSBS and NL of self-etch universal adhesives, but this influence was material-dependent.

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1.   Introduction

Several materials have been used to restore the ideal anatomy of severely damaged, fractured or extensively carious teeth, especially those with compromised resistance and retention form, prior to their preparation for indirect restorations. Resin-based core buildup materials are often used for this purpose in clinical dental practice [1–3]. Different materials such as amalgam, glass-ionomer cements, resin cements, flowable composite materials and regular viscosity composite materials, have been studied and suggested as core buildup materials in the literature. These materials present different characteristics regarding their strength and resistance to fracture, as well as their handling [4,5].

In spite of the variety of materials available for core buildup, core buildup resin composite materials in combination with an adhesive system are currently recommended. These materials have become very popular because they can be bonded to the remaining tooth structure to provide resistance and retention for the final restoration [1].

Core buildup resin composite materials are available in self-, light- and dual-cured formulations [6,7]. Dual-cured core buildup resin composite were developed in order to overcome limitations of self- and light-cured materials, by combining chemically- and light-induced polymerization initiation. Dual-cured materials contain both an oxidation-reduction (also known as redox) initiator system and photoinitiators [8]. Polymerization is mainly initiated by light activation in the superficial layers of the resin composite to achieve initial hardening, followed by chemical activation in deeper layers where the light irradiation is severely attenuated [8,9].

More recently, buildup resin composite materials have been used with simplified self-etch adhesive systems following the respective manufacturers' instructions. While the bonding between adhesives and dentin is not reliable [10,11], this shortcoming is even more pronounced for simplified adhesives [12,13]. In addition, simplified adhesives may be chemically incompatible with dual-cured core buildup resin composites [9,14]. Residual acidic monomers in the underlying oxygen inhibited layer formed by simplified adhesive systems with pH < 3 deactivate the initiator component (aromatic tertiary amine) inhibiting the polymerization reaction of resin composites that are initiated via peroxide-amine binary redox catalysts, resulting in weak polymerization [14–17], such as self- and dual-cured core buildup resin composites. Another mechanism that may account for the incompatibility between simplified self-step adhesives and dual- or self-cured resin composites is the presence of a hypertonic environment in the oxygen inhibited layer that activates osmotic fluid transport through the permeable adhesive layer [15].

In order to circumvent the potential incompatibility between simplified adhesives and dual-cured buildup resin composites, and ensure complete polymerization in deeper parts of the preparation, some light-cured simplified adhesives are currently paired with a self-curing activator, usually composed of arylsulfinate salts [18,19]. Nevertheless, it has been reported that the bonding ability of self-cured core buildup resin composites depends on the chemical composition of the individual adhesive system, as the very low or no measurable bonding to dentin obtained with incompatible materials may result in premature restoration failure [20–22]. In 1999 it was reported that bonding failures occurred when self-cured buildup resin composites were bonded with simplified adhesive systems. It was suggested that this phenomenon might be related to the adhesive's chemistry, specifically a low pH [22]. Unfortunately, this chemical incompatibility is not always reversed by adding a self-cured activator to the adhesive [23]. Moreover, the addition of an activator may not be effective for some specific adhesive compositions as the activator may dilute the adhesive in such an extent that it negatively affects adhesion [24].

Current universal adhesives are essentially simplified one-step self-etch adhesives with pH varying from 1.6 to 3.2 [25,26]; therefore they share most disadvantages with older one-step self-etch adhesives. In fact, universal adhesives may also be incompatible with self-cured and dual-cured buildup resin composites. To the extent of authors' knowledge, this potential compatibility has not been thoroughly investigated.

Thus, the purpose of the study was to evaluate the microshear bond strengths and nanoleakage of adhesive/core buildup resin composites as influenced by (1) curing protocols and (2) water storage. The following null hypotheses were tested: (1) bond strengths and nanoleakage do not change when the adhesive/core buildup material is bonded using different curing protocols, and (2) bond strengths and nanoleakage do not change when the adhesive/core buildup material is stored in water for 6 months.

2.   Material and methods

2.1.   Teeth preparation and bonding procedures

Fifty-five extracted and caries-free human third molars were used. The teeth were collected after obtaining the patients' informed consent under a protocol approved by the Ethics Committee Review Board of the local university. The teeth were disinfected in 0.5% chloramine, stored in distilled water, and used within 6 months after extraction. In each tooth, an occlusal cavity (4 mm × 4 mm) was prepared with the pulpal floor extending approximately 4 mm into dentin. The roots of all teeth were sectioned at the cementum–enamel junction. The crowns were then sectioned across the long axis of the teeth to obtain four dentin specimens (buccal, lingual, and proximals) (Fig. 1). Two-hundred twenty dentin specimens (N = 220) originating from 55 teeth were ground wet with #600-grit SiC paper for 60 s each and used for evaluation of the bond strength (n = 165) and nanoleakage (n = 55).

2.2.   Experimental design

The dentin specimens were randomly assigned into 11 experimental conditions (n = 20 dentin specimens; 15 to μSBS, 5 to nanoleakage) according to two independent variables: (1) *Adhesive (self-etch mode)/composite buildup material system*—All-Bond Universal/Core Flo DC, used as light-cured system control (ABU, Bisco Inc., Schaumburg, IL, USA), Clearfil SE Bond/Clearfil DC Core Plus, used as dual-cured system control (CSE, Kuraray Noritake Dental Inc., Tokyo, Japan), Clearfil
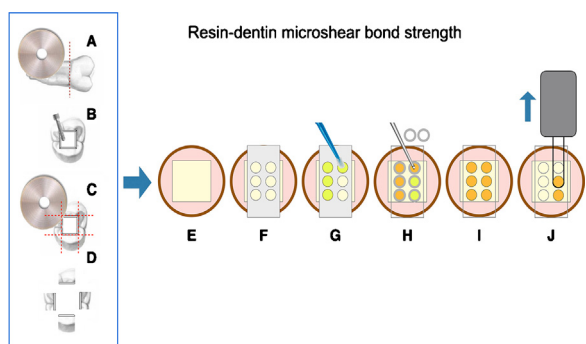
Fig. 1 – Schematic drawing showing specimen preparation and testing. (A) The roots of all teeth were sectioned at the cementum–enamel junction. After cavity preparation (B), the crowns were sectioned in two perpendicular directions across the long axis of the tooth (C) to produce four dentin specimens (buccal, lingual, and proximals; D). In (E) each dentin specimen was mounted on a PVC ring filled with acrylic resin (displaying the dentin surface on the top of the cylinder); (F) a perforated double-faced adhesive tape was then attached to the dentin specimens to delimit the bonding area. After adhesive application and light curing (G), Tygon tubes were adapted to the dentin surface (H), and each lumen was filled with core buildup resin composite and polymerized accordingly (H). After each storage time Tygon tubes and adhesive tapes were removed, leaving the bonded core buildup resin composite cylinders on the dentin surface (I). Each tooth was placed in a jig and assembled in a universal testing machine for microshear bond strength testing using an orthodontic-loop around the core buildup resin composite specimens (J).

Universal Bond/Clearfil DC Core Plus (CFU, Kuraray Noritake Dental Inc., Tokyo, Japan), Prime&Bond Elect/FluoroCore 2+ (PBE, Dentsply Sirona, Milford, DE, USA), and One Coat 7 Universal/ParaCore (OCU, Coltene/Whaledent AG, Altstätten, Switzerland); (2) *Curing protocol*—For CFU, PBE and OCU, three curing protocols were used: light-cured mode [LC], dual-cured mode [DC] and self-cured mode [SC]; and (3) *Storage time*—tested immediately [24 h] or after 6 months of storage [6 m] in distilled water.

2.3. Dentin microshear bond strength (μSBS)

Polyvinyl chloride (PVC) rings were filled with acrylic resin (AutoClear, DentBras; Pirassununga, São Paulo, Brazil). The specimens were embedded into the acrylic resin protruding 3 mm from the PVC ring. The delimitation of the bonding area was performed according to Shimaoka et al. [27]. Six perforations with an internal diameter of 0.8 mm were made in an acid-resistant double-faced adhesive tape (Adelbras Ind. e Com. Adesivos Ltda, SP, Brazil) with a Hygenic Ainsworth-style rubber dam punch (Coltène/Whaledent AG, Altstätten, Switzerland). This adhesive tape was then attached to the dentin specimens (Fig. 1). All specimens were randomized in block into different groups (www.sealedenvelope.com) prior to the application of the adhesive. A person not involved

in the research protocol performed the randomization using computer-generated tables.

A single operator performed all bonding procedures under a 10× magnification loupe. The adhesive systems were used in self-etch mode according to the following (Table 1): All-Bond Universal, light-cured mode (ABU/LC), as a LC control; Clearfil SE Bond, dual-cured mode (CSE/DC), as a DC control; Clearfil Universal Bond, light-cured mode (CFU/LC); Clearfil Universal Bond, dual-cured mode (CFU/DC); Clearfil Universal Bond, self-cured mode (CFU/SC); Prime&Bond Elect, light-cured mode (PBE/LC); Prime&Bond Elect, dual-cured mode (PBE/DC); Prime&Bond Elect, self-cured mode (PBE/SC); One Coat 7 Universal, light-cured mode (OCU/LC); One Coat 7 Universal, dual-cured mode (OCU/DC); One Coat 7 Universal, self-cured mode (OCU/SC).

After the application of the adhesive system, six polyethylene transparent Tygon tubes (Tygon Medical Tubing Formulations 54-HL, Saint Gobain Performance Plastics, Akron, OH, USA), with the same internal diameter of the perforations (0.8 mm) and a height of 0.5 mm were positioned over the double-faced tape, ensuring that their lumen coincided with the circular areas exposed by the perforations. The composite buildup material for each adhesive system was carefully packed inside each tube, and a clear Mylar matrix strip was placed over the filled Tygon tube and pressed gently into place (Fig. 1). The composite buildup material was light-cured according to the respective manufacturer's instructions (Table 1) using a LED light-curing unit set at 1200 mW/cm^2 (Radiical, SDI Limited, Bayswater, Victoria, Australia). A radiometer (Demetron L.E.D. Radiometer, Kerr Sybron Dental Specialties, Middleton, WI, USA) was used to check the light intensity every 5 specimens. These procedures were carried out under magnifying loupes [28].

After storage of the specimens in distilled water for 24 h at 37 °C, the Tygon tubes and the double-faced adhesive tape were carefully removed with a blade, exposing the composite buildup cylinders. Each specimen was examined under a loupe at 10× magnification. The composite buildup cylinders that originated from the same dentin specimen were randomly divided and assigned to be tested immediately [24 h] or after 6 months of storage [6 m] in distilled water at 37 °C. The storage solution was not changed and its pH was monitored monthly. The specimens were attached to a shear-testing fixture (Odeme Biotechnology, Joaçaba, SC, Brazil), and were tested immediately in a universal testing machine (Kratos IKCL 3-USB, Kratos Equipamentos Industriais Ltda, Cotia, SP, Brazil).

Each specimen was positioned onto the universal testing machine and a thin orthodontic wire (0.2 mm diameter) was looped around the base of each composite buildup cylinder (Fig. 1). The orthodontic wire contacted the composite buildup cylinder in half of its circumference. The setup was kept aligned (composite buildup cylinder-dentin interface, the wire loop and the center of the load cell) to ensure the correct orientation of the shear forces [29]. The crosshead speed was set at 1 mm/min until failure. The μSBS (MPa) were calculated by dividing the load at failure by the surface area (mm^2). The failure mode was classified as cohesive ([C] failure exclusively within dentin or composite buildup material), adhesive ([A] failure at the resin–dentin interface), or mixed ([M] failure

Table 1 – Adhesive and core buildup resin composite system (batch number), composition[a] and application mode.

Adhesive and core buildup (batch number) and pH[b]	Composition[a]	Self-etch, light cure mode (SE-LC)	Self-etch, dual cure mode (SE-DC)	Self-etch, self cure mode (SE-SC)
All-Bond Universal—ABU (1500003086) pH = 2.5–3.0	Adhesive: 10-MDP, Bis-GMA, HEMA, ethanol, water, initiators	1. Apply two separate coats of adhesive, scrubbing the preparation with a microbrush for 10–15 s per coat. Do not light cure between coats 2. Evaporate excess solvent by thoroughly air-drying with an air syringe for at least 10 s, there should be no visible movement of the material. The surface should have a uniform glossy appearance 3. Light cure for 10 s at 1200 mW/cm^2 4. Apply core buildup and light cure for 40 s at 1200 mW/cm^2		
Core Flo DC (1500003885)	Resin matrix: Bis-GMA, ethoxylated Bis-GMA, TEGDMA Filler: glass filler, fumed silica, amorphous silica			
Clearfil SE Bond—CSE Primer (01249A) pH = 1.9	Primer: 10-MDP, HEMA, dl-camphorquinone, hydrophilic, aliphatic dimethacrylate, N,N-diethanol-p-toluidine, water.		1. Apply primer with a disposable brush tip. Leave it in place for 20 s 2. Evaporate the volatile ingredients with a mild oil-free air stream 3. Dispense each one drop of bond and activator into a well of the dispensing dish and mix them with the applicator brush 4. Apply the mixture and rub it for 10 s 5. After application, make the bond film as uniform as possible using a gentle oil-free air stream 6. Light cure for 10 s at 1200 mW/cm^2 7. Apply core buildup and light cure for 20 s at 1200 mW/cm^2	
Adhesive (01887A) pH = 2.8	Adhesive: 10-MDP, HEMA, Bis-GMA, hydrophobic aliphatic dimethacrylate, dl-camphorquinone, N,N-diethanol-p-toluidine, colloidal silica			
Clearfil DC Activator—(7F0002)	Activator: ethanol, catalysts, accelerators			

Clearfil DC Core Plus (3R0147)	A Paste: Bis-GMA, hydrophobic aliphatic dimethacrylate, hydrophilic aliphatic dimethacrylate, hydrophobic aromatic dimethacrylate, silanated barium glass filler, silanated colloidal silica, colloidal silica, dl-camphorquinone, initiators, pigments B Paste: triethyleneglycol dimethacrylate, hydrophilic aliphatic dimethacrylate, hydrophobic aromatic dimethacrylate, silanated barium glass filler, silanated colloidal silica, aluminum oxide filler, accelerators			
Clearfil Universal Bond—CFU (C50002) pH = 2.3	Adhesive: 10-MDP, Bis-GMA, HEMA, di-camphorquinone, hydrophilic aliphatic dimethacrylate, silane coupling agent, colloidal silica and accelerators, ethanol, water	1. Apply bond and rub it for 10 s 2. Dry by blowing mild air for 5 s 3. Light-cure for 10 s at 1200 mW/cm^2 4. Apply core buildup and light cure for 20 s at 1200 mW/cm^2	1. Dispense each one drop of bond and activator into a well of the dispensing dish and mix them with the applicator brush 2. Apply the mixture and rub it for 10 s 3. Dry by blowing mild air for 5 s 4. Light-cure for 10 s at 1200 mW/cm^2 5. Apply core buildup and light cure for 20 s at 1200 mW/cm^2	1. Dispense each one drop of bond and activator into a well of the dispensing dish and mix them with the applicator brush 2. Apply the mixture and rub it for 10 s 3. Dry by blowing mild air for 5 s 4. Apply core buildup and wait for 20 min 5. Light cure for 20 s at 1200 mW/cm^2
Clearfil DC Activator—(7F0002) Clearfil DC Core Plus (3R0147)	Activator: ethanol, catalysts, accelerators A Paste: Bis-GMA, hydrophobic aliphatic dimethacrylate, hydrophilic aliphatic dimethacrylate, hydrophobic aromatic dimethacrylate, silanated barium glass filler, silanated colloidal silica, colloidal silica, dl-camphorquinone, initiators, pigments B Paste: triethyleneglycol dimethacrylate, hydrophilic aliphatic dimethacrylate, hydrophobic aromatic dimethacrylate, silanated barium glass filler, silanated colloidal silica, aluminum oxide filler, accelerators			

Table 1 (Continued)

Adhesive and core buildup (batch number) and pH[b]	Composition[a]	Self-etch, light cure mode (SE-LC)	Self-etch, dual cure mode (SE-DC)	Self-etch, self cure mode (SE-SC)
Prime&Bond Elect—PBE (130202) pH = 2.5	Adhesive: Mono-, di- and trimethacrylate resins; PENTA Diketone; organic phosphine oxide; stabilizers; cetylamine hydrofluoride; acetone; water	1. Apply generous amount of adhesive using microbrush. Agitate for 20 s 2. Gently dry with clean, dry air from a dental syringe for'at least 5 s. Surface should'have a uniform glossy appearance 3. Light cure for 10 s at 1200 mW/cm^2 4. Apply core buildup and light cure for 20 s at 1200 mW/cm^2	1. Dispense each one drop of bond and activator into a clean plastic mixing well mix them for 2 s with a clean unused brush tip 2. Apply generous amount of mixed. Agitate the applied adhesive/activator mixture for 20 s 3. Gently dry with clean, dry air from a dental syringe for'at least 5 s. Surface should'have a uniform glossy appearance 4. Light cure for 10 s at 1200 mW/cm^2 5. Apply core buildup and light cure for 20 s at 1200 mW/cm^2	1. Dispense each one drop of bond and activator into a clean plastic mixing well mix them for 2 s with a clean unused brush tip 2. Apply generous amount of mixed. Agitate the applied adhesive/activator mixture for 20 s 3. Gently dry with clean, dry air from a dental syringe for'at least 5 s. Surface should 'have a uniform glossy appearance 4. Apply core buildup and wait for 20 min 5. Light cure for 20 s at 1200 mW/cm^2
Self Cure Activator (141222)	Activator: UDMA, HEMA, catalyst, photoinitiators, stabilisers, acetone, water			
FluoroCore 2+ (150608)	Urethane dimethacrylate; di- & tri-functional methacrylates; barium boron fluoroaluminosilicate glass; camphorquinone (CQ) photoinitiator; photoaccelerators; silicon dioxide; benzoyl peroxide			

One Coat 7 Universal—OCU (G07542) pH = 2.8	Adhesive: 10-MDP, methacrylated polyacrylic acid, other methacrylates, photoinitiators, ethanol, water	1. Dispense a drop of adhesive and rub it onto the dentin with a disposable dental brush for 20 s 2. Blow gently with oil-free compressed air for 5 s 3. Light cure for 10 s at 1200 mW/cm^2 4. Apply core buildup and light cure for 20 s at 1200 mW/cm^2	1. Dispense a drop of adhesive and rub it onto the dentin with a disposable dental brush for 20 s 2. Blow gently with oil-free compressed air for 5 s 3. Dispense one new drop of adhesive and one drop of activator into a dispensing well and mix it well with a clean disposable brush (approx. 5–10 s) 4. Apply the mixed bond using a disposable brush onto the dentin 5. Gently dry for 5 s using oil-free compressed air 6. Light cure for 10 s at 1200 mW/cm^2 7. Apply core buildup and light cure for 20 s at 1200 mW/cm^2	1. Dispense a drop of adhesive and rub it onto the dentin with a disposable dental brush for 20 s 2. Blow gently with oil-free compressed air for 5 s 3. Dispense one new drop of adhesive and one drop of activator into a dispensing well and mix it well with a clean disposable brush (approx. 5–10 s) 4. Apply the mixed bond using a disposable brush onto the dentin 5. Gently dry for 5 s using oil-free compressed air 6. Apply core buildup and wait for 20 min 7. Light cure for 20 s at 1200 mW/cm^2
One Coat 7.0 Activator—(G46401)	Activator: ethanol, water, activator			
ParaCore (G26422)	Resin matrix: Bis-GMA, TEGDMA, UDMA; Filler: fluoride, barium glass, amorphous silica (68 wt%, 0.1–5 mm)			

[a] 10-MDP = methacryloyloxydecyl dihydrogen phosphate; Bis-GMA = Bisphenol A diglycidylmethacrylate; HEMA = 2-hydroxyethyl methacrylate; PENTA = dipentaerythritol penta acrylate monophosphate; TEGDMA = triethyleneglycol dimethacrylate; UDMA = urethane dimethacrylate.

[b] [46,47,59].

at the resin–dentin interface that included cohesive failure of the neighboring substrates). The failure mode analysis was performed under a stereomicroscope at 100× magnification (Olympus SZ40, Tokyo, Japan).

2.4. Nanoleakage evaluation (NL)

Five randomized dentin specimens for each condition (one per tooth) were used for nanoleakage evaluation. A single operator performed all bonding procedures according to Table 1. Subsequently each dentin specimen was divided in two parts, which were randomly assigned to be tested immediately [24 h] or after 6 months of storage [6 m] in distilled water at 37 °C. All composite buildup material-dentin specimens were coated with two layers of nail varnish applied up to within 1 mm of the bonded interfaces. The composite buildup material-dentin specimens were immersed in 50 wt% ammoniacal silver nitrate solution in total darkness for 24 h, rinsed thoroughly in distilled water, and immersed in photo developing solution for 8 h under a fluorescent light to reduce silver ions into metallic silver grains within voids along the bonded interface [30].

Specimens were polished with SiC paper of decreasing abrasiveness up to 1200-grit and 1 and 0.25 μm diamond pastes (Buehler Ltd., Lake Bluff, IL, USA). Then, they were ultrasonically cleaned in distilled water, mounted on aluminum stubs, air-dried and gold sputter coated (MED 010, Balzers Union, Balzers, Liechtenstein). The interfaces were observed in a scanning electron microscope (SEM) in backscattered mode at 12 kV (VEGA 3 TESCAN, Shimadzu, Tokyo, Japan).

With the objective of standardizing image acquisition, five micrographs were taken for each of the five specimens. The first micrograph was taken in the center of the composite buildup material-dentin specimen. The other four micrographs were obtained 0.3 mm and 0.6 mm to the left and right of the first one. One dentin specimen per tooth was evaluated and a total of five dentin specimens were used for each experimental condition, a total of 25 images were evaluated per group [31]. A technician who was blinded to the experimental conditions under evaluation obtained all micrographs. The relative percentage of nanoleakage within the adhesive and hybrid layer areas was measured in all micrographs using ImageJ software (National Institutes of Health, Bethesda, MD, USA) [32].

2.5. Statistical analysis

The μSBS of all specimens with adhesive/mixed failure mode from the same dentin specimens were averaged for statistical purposes. Similarly, the same procedure was performed for the NL evaluation, so that the experimental unit in this study was the dentin specimens. Specimens with cohesive and premature failures were not included in the data analysis. Data from μSBS and NL were analyzed separately. Two statistical analysis were performed: 1- data from all groups were analyzed using two-way ANOVA (*adhesive/composite buildup system* vs. *storage time*) and; 2- data from each adhesive were analyzed using two-way ANOVA (*curing protocol vs. storage time*). A Tukey's post hoc test at $\alpha = 0.05$ was used for both tests.

3. Results

3.1. Dentin microshear bond strength (μSBS)

Ninety bonded cylinders were tested for each group, forty-five in each evaluation time. The majority of specimens showed adhesive/mixed failures (Table 2). When the 24 h results were evaluated, the LC control group (ABU) showed similar mean μSBS to those of CFU/LC and PBE/LC (p > 0.05; Table 3). However, OCU/LC showed statistically significant higher mean μSBS when compared with ABU (p < 0.001; Table 3). In DC mode, the control group CSE showed statistically significant higher mean μSBS compared to all universal adhesives in DC mode (p < 0.001; Table 3).

When each universal adhesive was compared in the different curing protocols at 24 h, only PBE/LC resulted in statistically significant lower mean μSBS compared with the respective DC and SC mode (p < 0.001; Table 3). CFU and OCU were not significantly affected by curing protocol (p > 0.05; Table 3).

After 6-month of water storage, no significant changes in mean μSBS were observed for either control group (ABU and CSE) when compared with the respective 24 h results (p > 0.05; Table 3). No significant changes were observed for CFU and PBE in any curing protocol when compared with their respective 24 h values (p > 0.05; Table 3). For OCU only a significantly decrease in mean μSBS was observed in DC mode when 6 m results were compared with the 24 h results (p < 0.0001; Table 3).

3.2. Nanoleakage (NL)

When 24 h results were evaluated, the control group for LC mode (ABU) showed mean %NL similar to those of CBU/LC and OCU/LC (p > 0.05; Table 4). However, PBE/LC showed statistically significant higher mean %NL compared with ABU (p < 0.001; Table 4). The DC control group (CSE) showed similar mean %NL when compared to CFU/DC (p > 0.05; Table 4). However, CSE resulted in statistically higher mean %NL when compared to OCU/DC and statistically lower mean %NL when compared to PBE/DC (p < 0.001; Table 3). Representative SEM micrographs of each group are shown in Fig. 2.

When each universal adhesive was compared in the different curing protocols at 24 h, CFU in both LC and SC mode resulted in statistically significant lower mean %NL when compared with CFU/DC (p < 0.001; Table 3). PBE/SC resulted in statistically significant lower mean %NL when compared with PBE/LC and PBE/DC (p < 0.001; Table 3). OCU/LC and OCU/DC resulted in statistically significant lower mean %NL when compared with OCU/SC (p < 0.001; Table 3).

After 6-month of water storage, both control groups (ABU and CSE), as well as, CFU/LC and CFU/SC, resulted in statistically significant increase of mean %NL when compared with their respective 24 h results (p < 0.0001; Table 4). There were no signs of increase of nanoleakage for PBE and OCU (p > 0.05; Table 4), although PBE showed the worst (highest) %NL (p > 0.05; Table 4). Representative SEM micrographs of each group are shown in Fig. 3.

Table 2 – Number of specimens according to fracture mode for all experimental groups.[a]

| | Immediate | | | | | | | | | 6-month water storage | | | | | | | | |
| | LC | | | DC | | | SC | | | LC | | | DC | | | SC | | |
	A	C	M	A	C	M	A	C	M	A	C	M	A	C	M	A	C	M
All-Bond Universal	25	0	20							17	3	25						
Clearfil SE Bond				40	1	4							35	0	10			
Clearfil Universal	22	1	22	27	0	18	25	2	18	19	3	23	23	3	19	20	3	22
Prime&Bond Elect	35	0	10	31	2	12	27	2	16	27	3	15	18	2	25	20	3	22
One Coat Bond	33	3	9	29	2	14	22	0	23	26	0	19	22	1	22	18	8	19

[a] A = failure at the resin–dentin interface; C = cohesive (failure exclusively within dentin or core buildup) or; M = mixed (failure at the resin–dentin interface that included cohesive failure of the neighboring substrates).

Table 3 – Mean and standard deviation of microshear bond strength (MPa) to dentin for each experimental condition.[*],[**]

| | Immediate | | | 6-month water storage | | |
	LC	DC	SC	LC	DC	SC
All-Bond Universal	17.0 ± 3.5 D			15.6 ± 2.5 D,E		
Clearfil SE Bond		25.9 ± 2.0 A			25.6 ± 1.1 A	
Clearfil Universal	18.4 ± 1.9 C,D a,b	20.8 ± 2.6 B a	24.1 ± 2.5 A a	15.9 ± 2.9 D,E b	19.9 ± 2.3 B,C a	21.3 ± 2.4 B a
Prime&Bond Elect	15.8 ± 2.3 D,E d,e	21.3 ± 1.0 B c	22.6 ± 2.8 B c	13.9 ± 1.8 E e	21.0 ± 3.3 B c	18.9 ± 1.6 C,D c,d
One Coat Bond	20.4 ± 1.8 B,C f	20.4 ± 1.1 B,C f	23.2 ± 1.2 A,B f	20.4 ± 1.0 B,C f	16.2 ± 3.6 D,E g	23.0 ± 1.5 A,B f

[*] Different superscript letters mean statistically significant differences between groups (columns) (Tukey test, $p < 0.05$).
[**] Different lowercase letters mean statistically significant differences for each adhesive system (rows) (Tukey test, $p < 0.05$).

Table 4 – Mean and standard deviation of nanoleakage (NL) in dentin for each experimental condition.[*],[**]

| | Immediate | | | 6-month water storage | | |
	LC	DC	SC	LC	DC	SC
All-Bond Universal	7.9 ± 2.0 A,B			36.5 ± 4.7 E,F		
Clearfil SE Bond		15.3 ± 2.1 C			30.3 ± 5.5 E	
Clearfil Universal	3.0 ± 3.7 A a	13.0 ± 3.4 C b	4.0 ± 7.9 A,B a	18.9 ± 3.4 D b	16.2 ± 3.3 C,D b	19.8 ± 4.4 D,E b
Prime & Bond Elect	47.6 ± 10.9 F d	40.8 ± 7.5 F d	26.3 ± 2.8 E c	52.2 ± 8.8 F d	47.4 ± 6.3 F d	32.9 ± 7.2 E c
One Coat Bond	0.9 ± 1.8 A f	5.6 ± 4.3 A,B f,g	18.2 ± 3.5 D h	1.4 ± 1.0 A f	10.2 ± 2.3 A B g	18.5 ± 1.5 D h

[*] Different superscript letters mean statistically significant differences between groups (columns) (Tukey test, $p < 0.05$).
[**] Different lower case letters mean statistically significant differences for each adhesive system (rows) (Tukey test, $p < 0.05$).

4. Discussion

CSE was used as the DC control as Raimondi et al. [33] reported higher shear bond strengths for this adhesive in DC mode compared to five universal adhesives also in DC mode. The 10-MDP molecules and the hydrophobic resin layer associated with CSE make this material a golden standard for self-etch adhesives [34,35]. CSE resulted in the highest mean bond strengths when compared with all universal adhesives in the same DC polymerization mode. CSE is the only two-step adhesive system evaluated in the present study, in contrast to universal adhesives that are considered one-step self-etch adhesives. The second bottle of CSE is a solvent-free hydrophobic resin coating. On the contrary, simplified SE adhesives do not have a hydrophobic resin coating, usually forming very thin coatings [36,37]. The thin coatings may be oxygen inhibited, resulting in a poorly polymerized adhesive layer [38]. The use of a less hydrophilic resin coating can be an option for increasing the degree of conversion of

the adhesive layer [39,40]. Simplified SE adhesives are considered permeable membranes after polymerization [41]. This can be explained due to the presence of water-attracting hydrophilic domains [42] and interconnecting water-filled channels (water–trees) within the polymerized adhesives, permitting water to move from the underlying dentin through the adhesive [30].

This study showed no significant differences for PBE and OCU when the DC mode was compared to the SC mode at 24 h. The results for PBE/DC and PBE/SC are in agreement with those of Raimondi et al. [33]. Also, when CFU was used in SC mode, the mean bond strengths increased compared to those of DC mode at 24 h. This outcome was unexpected, since, as described in the introduction section, there is a known incompatibility between the acid-containing simplified self-etch adhesive and DC core buildup resin composites [9,14]. This is because the adhesives' residual acidic monomers in the oxygen inhibition layer can deactivate the tertiary amines (base) that catalyze the polymerization of SC resin compos-
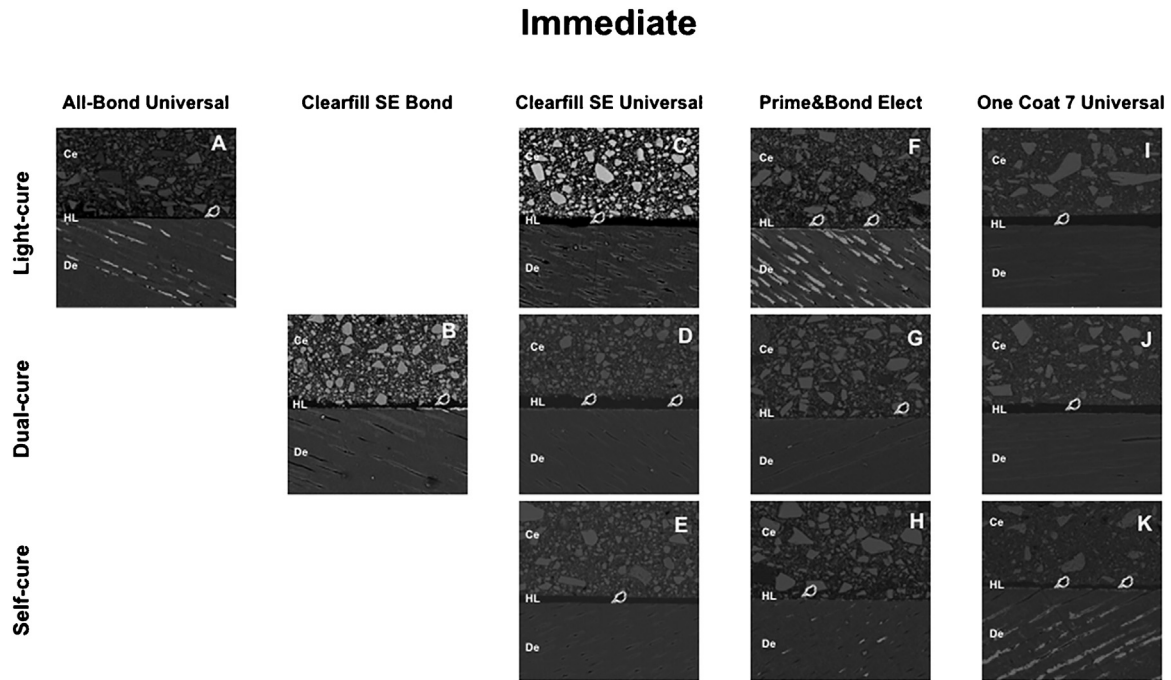
Immediate

Fig. 2 – Representative SEM micrographs of the resin-dentin interfaces of different experimental groups in the immediate time. Higher percentage of silver penetration was obtained for Prime&Bond Elect adhesive groups (F, G and H), mainly in the hybrid layer (white pointers). (Ce = core buildup; De = dentin; HL = hybrid layer).
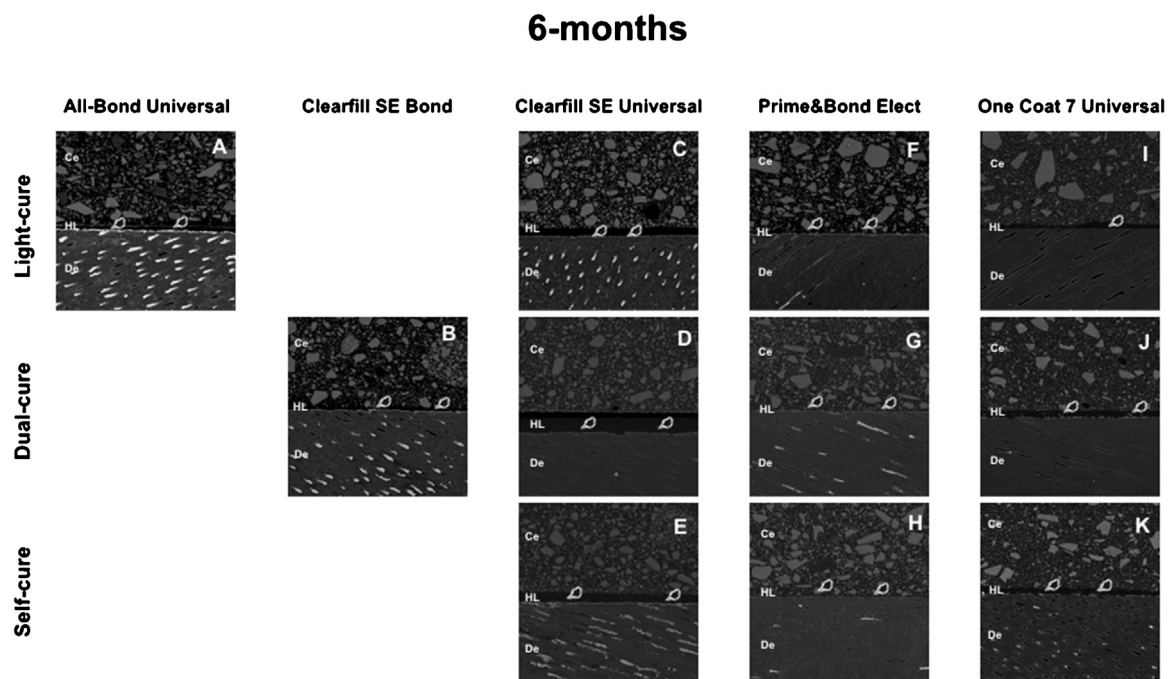
6-months

Fig. 3 – Representative SEM micrographs of the resin-dentin interfaces of different experimental groups after 6-month water storage. Higher percentage of silver penetration was shown for Prime&Bond Elect adhesive groups (F, G and H), mainly in the hybrid layer (white pointers), as well as, for the control groups (Ce = core buildup; De = dentin; HL = hybrid layer).

ites/cements [14–17]. However, the previous literature is based on older simplified self-etch adhesive systems. More recently, different manufacturers have developed simplified adhesives to be more hydrophobic (i.e., less hydrophilic) and less water permeable [43,44]. The inclusion of 10-MDP makes universal adhesives more hydrophobic than their predecessors, as this monomer is quite hydrophobic due to its long carbonyl chain [45].

A recent paper evaluated the role of the hydrophilicity of adhesives on the compatibility between simplified SE adhesives and a DC resin cement that was light cured or chemically cured [46]. The authors evaluated a self-etch adhesive with different degrees of hydrophilicity (10–30% of hydroxyl-etil methacrylate—HEMA), but with the same pH. In comparison two simplified commercial SE adhesives, ABU and Optibond All-in-One (Kerr) were also evaluated. The results showed that there was no incompatibility between the adhesive and the LC or SC resin cement for more hydrophobic adhesives, as well as for the universal adhesive evaluated (ABU). However, for more hydrophilic adhesives, there were significant drops in mean bond strengths (80–93%) when the adhesive was associated with a SC resin cement [46]. We hypothesize that the unexpected similar mean bond strengths obtained with different curing modes of both OCU and CFU in our study may be a consequence of their reduced hydrophilicity, in line with the findings of Chen and Suh for ABU [46].

Unfortunately, the exact composition of each universal adhesive is proprietary information. However, the SDS of OCU indicates that the amount of HEMA varies between 5–10%. For CFU, this amount varies between 10–35%, while for PBE this amount is lower than 20%. Future studies need to evaluate this variable for several commercial universal adhesives. It worth mentioning that we selected ABU as the LC control, mainly because ABU is considered a less hydrophilic universal adhesive, containing 10-MDP in its composition [15]. Also, according to the respective manufacturer's instructions, ABU does not require a separate self-cured activator with SC or DC resin composite materials, due to its fairly high pH of 3.2, compared to other universal adhesives [25,46,47].

OCU showed higher mean bond strengths compared to ABU at 24 h. The lower amount of HEMA may have been responsible for the higher bond strengths of OCU in LC more compared to other adhesives in the same curing mode (ABU and PBE). As HEMA has been shown to inhibit interfacial nano-layering of the molecule 10-MDP with calcium in hydroxyapatite [48], the chemical bonding ability of OCU may have been stronger than that of other adhesives with a higher HEMA concentration. HEMA promotes water uptake and hydrolysis at the adhesive interface, affecting the durability of the bond [12,49].

For PBE/LC mode, previous studies have reported lower dentin bond strengths compared to other universal adhesives [33,50,51]. PBE contains acetone as solvent, which has been known to prevent direct access of air to the surface of the resin monomer [52], forming a deficient air-inhibited layer. Due to the rapid evaporation of acetone, acetone-based resin monomers do not form a uniform layer on the dentin surface and may not plug dentinal tubules [53]. The high technique-sensitivity of LC acetone-based adhesives may be responsible for poor long-term results [54,55]. Additionally, PBE is more hydrophilic as it does not contain 10-MDP. These factors might be responsible for the lower adhesion capability of PBE in LC mode.

The relatively lower mean uSBS of both PBE/LC and CFU/LC compared to their respective DC and SC modes (although not significant for CFU in the immediate time) may be a result of the low pH of both adhesives (pH = 2.5 for PBE, pH = 2.3 for CFU). As mentioned in the Introduction, residual acidic monomers in the underlying oxygen inhibited layer of acidic adhesive systems deactivate the aromatic tertiary amine initiator, inhibiting the polymerization reaction of resin composites that are initiated via peroxide-amine binary redox catalysts [14]. The addition of the self-curing activator may have reversed this reaction for both adhesives PBE and CFU in DC and SC mode, resulting in higher mean uSBS.

Additionally the %NL obtained with PBE, when polymerized in all curing modes, were significantly higher than the %NL of other adhesives tested, in line with the data previously reported by Luque-Martinez et al. [50]. Nanoleakage reveals the location of defects at the resin-dentin interface that may serve as pathways for degradation of resin-dentin bonds over time. Silver nitrate ions occupy nanometer-sized spaces around unprotected collagen fibrils, where resin failed to infiltrate or where residual water/solvent were not displaced by the adhesive resin [56]. As acetone present in the PBE composition evaporates faster than ethanol present in all others adhesives tested, the application of only one coat of PBE may have been insufficient to promote a complete resin infiltration inside the hybrid layer The higher amount of solvent in PBE (less than 50%) when compared to CFU (less than 20%) and OCU (between 35–40%) may also have contributed to this deficient infiltration. Additionally, PBE may be too hydrophilic, as it does not contain 10-MDP, as previously mentioned. This monomer is quite hydrophobic due to its long carbonyl chain [45]. For PBE, the increase in bond strengths and decrease of %NL from LC to SC may be related to the extended time that the adhesive was allowed to interact with hydroxyapatite in the SC mode (20 min), which may have increased its pH from hydroxyapatite buffering [57]. This same phenomenon may occur with CFU in terms of decreasing bond strengths. On the other hand, this was not observed for OCU. According to Hume [57], such reaction would be less relevant for adhesives with higher pH. The pH of OCU is 2.8 [Böhner R.—personal communication], which is slightly less acidic than the pH of CFU (pH = 2.3) and PBE (pH = 2.5) [47].

This same extended buffering reaction may have been responsible for decreased bond strengths for both PBE and CFU after 6-month water storage, as PBE and CFU showed lower mean bond strengths in SC mode when compared with the 24 h results. Although the higher buffering capacity of dentin and the high reactivity of H$^+$ insure that little H$^+$ diffuses through dentin [58], Carvalho et al. [59] observed the occurrence of a porous zone of mineralized dentin in the corresponding undemineralized sections of self-etch adhesives and the authors hypothesized that this occur due to the accumulation of acidic but non-polymerizable hydrolytic adhesive components in some acidic one-step adhesives. These non-polymerizable acidic monomers may continue to etch the underlying dentin after the formation of the hybrid layer being responsible for continuous demineralization of the surrounding dentin, mainly when stored in water [60].

In summary, the first null hypothesis was rejected, as the mean μSBS and %NL of universal adhesives varied according to the curing protocol used. The second null hypothesis was rejected, as mean μSBS and %NL of some universal adhesives varied after 6 m water storage.

5.   Conclusion

For self-etch universal adhesives, the addition of a self-cured activator and different curing protocols influenced μSBS and NL, but this influence was material dependent.

Conflict of interest

Author Jorge Perdigão has received compensation for speaking at events sponsored by Coltene. This relationship has been reviewed and managed by the University of Minnesota in accordance with its conflict of interest polices. All other authors certify that they have no proprietary, financial, or other personal interest of any nature or kind in any product, service and/or company presented in this article.

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