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Assessment of the initial and aged dentin bond strength of universal adhesives ☆

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ABSTRACT

This study evaluated the effect of mechanical loading on microtensile bond strengths (μTBS) of universal adhesives to dentin and quantified adhesive dentin penetration using micro-Raman spectroscopy. Human molars had occlusal dentin exposed and were allocated into eight groups: All-Bond Universal and Scotchbond Universal using etch-and-rinse and self-etch approaches, Adper Prompt L-Pop, Adper Single Bond Plus, Clearfil SE Bond, and Optibond FL. Following bonding procedures and build-ups, specimens were either stored in water at 37 °C for 24 h or mechanically loaded (50,000 cycles, 50 N) prior to μTBS test. Additional teeth were prepared for micro-Raman analysis of adhesive penetration and FE-SEM. Data were analyzed by two-way ANOVA and Tukey's post hoc test ($P < 0.05$). Mechanical loading had no deleterious effect on μTBS with the exception of Adper Prompt L-Pop. Incomplete infiltration of the demineralized dentin was noticed for adhesives using the etch-and-rinse approach and for Scotchbond Universal in the self-etch approach.

1. Introduction

Dentin adhesion may be facilitated by etch-and-rinse and self-etch strategies. The use of etch-and-rinse adhesives for dentin bonding is based on the concept developed by Dr. Fusayama in the late 1970s when it became apparent that removal of the smear layer resulted in better adhesion [1]. The first clinically successful materials using the etch-and-rinse strategy, namely the three-step etch-and-rinse adhesives, were developed in the 1990s. These were followed by the simplified two-step etch-and-rinse adhesives. Dentin adhesion with etch-and-rinse adhesives is accomplished through removal of the smear layer, acidic demineralization of the underlying dentin, and subsequent adhesive infiltration of the exposed collagen [2]. While etch-and-rinse adhesives are applied after dentin etching with 30–40% phosphoric acid, self-etch adhesives prepare the dentin substrate for adhesive resin application without the need for rinsing with water. Various one- or two-step self-etch formulations have been designed for the purpose of reducing the technique sensitivity associated with rinsing and drying steps. Some formulations have sought to enhance the durability of the adhesive interface by including resin monomers with chemical affinity for hydroxyapatite [3]. Materials of different pH values have been fabricated so as to enable better control of smear layer removal and superficial dentin demineralization as well as better adhesive infiltration [4].

Recently introduced "universal" or "multi-mode" dental adhesives may be applied following etch-and-rinse or self-etch strategies [5–10]. These materials have performed well in immediate dentin bond strength tests regardless of the application mode [6]. However, potential long-term stability of the adhesive interfaces created by these materials is unknown as data on aged specimens is not available. Short-term *in vitro* studies, where other self-etch adhesives have been used after phosphoric acid etching of dentin, have shown a decrease in bond strengths [11,12]. It has been reported that the interaction between self-etch adhesives and dentin might be affected by removal of hydroxyapatite from the substrate [13]. In addition, full infiltration of the exposed collagen by the adhesive resin after treatment of dentin with phosphoric acid has not been possible with current materials [14–17]. No data reporting the interaction of universal adhesives with phosphoric acid-etched dentin are available.

Components included in universal adhesive formulations – namely the phosphorus-containing monomers 10-methacryloyloxydecyl dihydrogen phosphate (MDP) [18] and dipentaerythritol

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pentaacrylate phosphoric acid ester (PENTA) [13], and polyalkenoic acid copolymer (Vitrebond Copolymer, 3M ESPE, St. Paul, MN, USA) [19,20] – may enhance adhesion to tooth structures and have been part of the composition of different materials for decades. Particular interest has been given to MDP, which has been the topic of several research projects evaluating its chemical interactions to dental structures [3,18,21–28]. Adhesives containing MDP and PENTA have performed well in long-term clinical trials [29–31]. However, scarce literature is available on universal adhesives using such monomers. The MDP-containing adhesive All-Bond Universal (Bisco, Inc., Schaumburg, IL, USA) may perform better in the etch-and-rinse mode than in the self-etch mode [8]. Another MDP-containing adhesive – namely Scotchbond Universal (3M ESPE) – has shown promising results in the etch-and-rinse and self-etch application modes [6,10]. It is important to note that Scotchbond Universal, in addition to MDP, also contains the Vitrebond Copolymer which has chemical affinity for hydroxyapatitie [32]. The positive short-term results for Scotchbond Universal have been corroborated in a clinical trial where retention rates of 100% and 94% after 18 months of clinical service in the etch-and-rinse and self-etch modes were shown, respectively [9]. Assessing the performance of these materials in well-designed clinical trials is essential as oral conditions challenge the integrity of the adhesive interface. However, supporting *in vitro* studies enable insight into the subtleties of the adhesive interface not otherwise possible.

Teeth are subjected to cyclic loading during chewing and swallowing. The repetitive forces may induce mechanical and chemical changes in dentin-adhesive bonds and, as a result, challenge the long-term performance of restorations. *In vitro* evaluation of universal adhesives with simulation of oral conditions may improve prediction of clinical performance. Several laboratory investigations have reported that accelerated aging through mechanical loading can challenge the dentin/adhesive interface [33–40]. Specimens in the present study were subjected to mechanical loading in an attempt to mimic one aspect of the oral environment [15–17,41,42]. It has also been observed that diffusion of the adhesive resin into the demineralized dentin, with consequent formation of a well infiltrated hybrid layer, is critical for adequate bonding [43]. Ideally, the interlocking of collagen and adhesive resin would be such that no fluid filled voids would be left in the hybrid layer [44]. It has been noted that micro-Raman analysis may help in determining the extent of adhesive infiltration. Numerous studies using micro-Raman analysis have shown that the extent of adhesive penetration of etch-and-rinse adhesive systems is less than the depth of dentin demineralization [14–17]. In contrast, self-etch adhesives have often been reported to penetrate to the extent of dentin demineralization [17,41,45]. No micro-Raman studies have been carried out to investigate the interaction between universal adhesives and dentin, aside from evaluation of degree of conversion [8,46–48].

The purpose of this study was to evaluate the effects of mechanical loading on microtensile bond strengths (μTBS) of universal adhesives to dentin and to quantify the levels of adhesive penetration into dentin using micro-Raman spectroscopy. The hypotheses tested were that: (1) mechanical loading negatively affects μTBS of universal adhesives and (2) universal adhesives incompletely infiltrate the demineralized dentin when applied in the etch-and-rinse mode.

2. Materials and methods

2.1. Specimen preparation

Eighty-eight intact human third molars were collected and stored in 0.5% Chloramine T solution at 4 °C until use. All teeth were embedded in epoxy resin (Buehler, Lake Bluff, IL, USA) and the occlusal third of the crowns were removed using a low-speed diamond saw (IsoMet 1000, Buehler) under running water. The coronal structure was sectioned perpendicular to the tooth long axis, approximately 2 mm apical to the level of the occlusal grooves. The exposed mid-coronal dentin surfaces were polished with #600-grit silicon carbide paper for 60 s to ensure a standardized smear layer [49]. The teeth were randomly assigned into eight groups and treated as detailed in Table 1. Table 1 shows manufacturers, compositions, and application protocols. To ensure minimal variation in depth of dentin demineralization, the same etchant was used in all etch-and-rinse groups (Scotchbond Etchant, 3M ESPE) [50].

After adhesive application, composite resin build-ups, using TPH[3] (Dentsply Caulk, Milford, DE, USA), were created by addition of three increments of approximately 2 mm each. Build-ups, which had a flat "occlusal" surface, were approximately 6 mm in height by 10 mm in width by 10 mm in depth. The build-ups were placed without the aid of any matrix and were centralized on the prepared dentin surface. Light-curing procedures were performed using a Smartlite IQ2 LED (Dentsply Caulk). Each increment was light-cured for 20 s. The light power intensity throughout the procedures was 1400 mW/cm^2.

2.2. Mechanical loading

Half of the bonded specimens in each adhesive group ($n=5$) were randomly assigned for mechanical loading after storage in distilled water at 37 °C for 24 h. Specimens were mounted in a custom holder, positioned within the chamber, and fixed to a chewing simulator (SD Mechatronik GmbH, Feldkirchen Westerham, Germany). There was a circumferential ~1 mm gap between the mounted specimens (embedded in epoxy resin) and holder that was filled with high-viscosity polyvinyl siloxane (Virtual, heavy body, Ivoclar Vivadent, Schaan, Liechtenstein) to simulate the periodontal ligament [51]. Specimens were then loaded under 50 N with the force centered on the composite resin build-up. The load was vertically delivered using a spherical stainless steel tip (5 mm in diameter) for 50,000 cycles with specimens immersed in distilled water, at room temperature.

2.3. μTBS testing and fracture mode analysis

Specimens were sectioned into beams either after 24 h of water storage at 37 °C following the bonding procedures or immediately after mechanical loading. Specimens were sectioned under water irrigation with a low-speed diamond saw (IsoMet 1000) to obtain beams with a cross-section area of ~0.9 mm^2 [52]. The dimensions of the specimens were recorded using a digital caliper (Mitutoyo, Tokyo, Japan). Nine beams from the central area (located above the dental pulp) of each tooth were used to reduce substrate-regional variability [53]. A total of 45 beams were tested in each group.

Beams were attached to a Ciucchi jig with cyanoacrylate glue (Super Glue Gel, Loctite, Westlake, OH, USA) and tested in μTBS using an EZ-Test machine (Shimadzu Corporation, Tokyo, Japan) at a crosshead speed of 1 mm/min [52]. The μTBS values were expressed in MPa by dividing the peak break force by the cross-sectional area of the bonded interface. A value of 0 MPa was assigned to specimens within the predetermined focus area (central area) that debonded during sectioning procedures. Each tooth specimen was considered as a unit with values from the nine beams tested for each specimen being averaged and used in the statistical analysis [54]. Assumptions of the equality of variances and the normal distribution of errors were checked with Kolmogorov–Smirnov and Shapiro–Wilk tests ($\alpha=0.05$). Data were examined with two-way ANOVA and Tukey's post hoc test. Statistical significance was set at the 95% confidence level. Statistical analysis was carried out with SPSS 14.0 software (SPSS, Inc., Chicago, IL, USA).