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Polymerization efficiency affects interfacial fracture toughness of adhesives

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ABSTRACT

Objective. To evaluate the effect of the kind and concentration of photo-initiator on the degree of conversion (DC) of adhesives on dentin/glass substrates and their mini-interfacial fracture toughness (mini-iFT) to dentin.
Methods. We tested the adhesive Clearfil S^3 Bond Plus and 4 derived experimental 'LUB' ('Leuven Univesity Bond') adhesives (all from Kuraray Noritake), namely 'LUB-CQ/amine_high', 'LUB-CQ/amine_low', 'LUB-TPO_high', and 'LUB-TPO_low', respectively containing 2.0 wt% camphorquinone (CQ) and 2.0 wt% EDMAB (amine), 0.35 wt% CQ and 0.35 wt% amine, 2.0 wt% TPO, and 0.35 wt% TPO. For DC, each adhesive was applied onto glass or dentin prior to being cured (Bluephase 20i; Ivoclar Vivadent: "high mode") for 10 s. DC was measured at 5 min, 10 min, 1 h, 24 h and 1 week using micro-Raman spectroscopy (SENTERRA; BrukerOptik). For mini-iFT, each adhesive was bonded to 320-grit SiC-paper ground dentin and covered with composite (Z100; 3 M ESPE). The restored teeth were cut in sticks (1.5 × 2.0 × 16 mm), after which a single-gradient notch was prepared at the adhesive-dentin interface using a 150-μm diamond blade. The micro-specimens were loaded until failure in a 4-point bending test and the mini-iFT in term of K_{QvM} was calculated.
Results. DC was higher on dentin than on glass. All adhesives were adequately polymerized at 1 week, except for LUB-TPO_low. DC at 5 min was significantly higher for LUB-TPO_high than for both CQ/amine-based adhesives. The highest and most reliable mini-iFT was measured for LUB-CQ/amine_high, despite its 5-min DC was relatively low. No correlation between DC and mini-iFT was found.
Significance. Curing of TPO-based adhesives is faster, but the dark cure of the CQ/amine-containing adhesives is more efficient. The differences in curing profiles do affect the mechanical properties of the resultant interfaces at dentin.

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1. Introduction

The photoinitiator kind and its concentration are of importance to dental adhesives and their eventual bonding effectiveness [1,2]. Reaching a sufficiently high degree of conversion (DC) not only contributes to the adhesive's bond strength and durability [2–4], but it is also associated with biocompatibility issues caused by monomer and/or photoinitiator elution [4–6]. Most commonly used photoinitiators in dental adhesives are camphorquinone combined with tertiary amine (CQ/amine) and diphenyl(2,4,6-trimethylbenzoyl)phosphine oxide (TPO).

We recently introduced a mini-interfacial fracture toughness (mini-iFT) test to assess bonding effectiveness of adhesives to tooth tissue [7,8]. This mini-iFT test enabled to discriminate adhesives better on their bonding effectiveness and, more importantly, the mini-iFT was shown to better reflect the mechanical strength of the actual interface than the today more commonly used micro-tensile bond-strength (μTBS) test [9]. A general consensus exists indeed in literature to preferentially use a fracture toughness approach to assess bonding effectiveness [10,11], despite the specimen-preparation methodology for interfacial fracture toughness testing is generally thought to be more technique-sensitive and labor-intensive. Several interfacial fracture toughness test protocols have been introduced, among which the most common are a 'short rod chevron notch' [12], a 'chevron-notched short bar' [13], a 'plane-strain chevron-notched short bar' [14], an 'interfacial fracture toughness related to the energy release rate' [15], a 'notchless triangular prism' [16] and a 'chevron notch beam' [17,18]. The major advantages of our newly developed mini-iFT test are that it is more accurate, more reproducible, less test dependent and that it reveals the interfacial properties better than a μTBS test. In addition, the mini-iFT test is less laborious and less time-consuming than a conventional interfacial fracture toughness. It has proven to be a valid and reliable laboratory test to determine bonding effectiveness to human dentin and enamel [7,8].

Since the mini-iFT test was shown to better assess the mechanical strength of the adhesive-tooth interface itself, we believed that this method would also suit well to investigate the effect of the kind and concentration of photoinitiator on the bonding effectiveness to dentin. Experimental adhesive formulations that differed for the kind of photoinitiator (CQ/amine versus TPO) and the photoinitiator concentration were tested. DC was measured over time when the adhesives were applied on dentin and glass in order to evaluate any correlation that may exist between DC and mini-iFT.

2. Materials and methods

2.1. Materials used

Five different adhesive formations, consisting of the commercially available adhesive Clearfil S^3 bond Plus (Kuraray Noritake, Tokyo, Japan) and 4 experimental derivatives (also provided by Kuraray Noritake), being referred to as 'LUB-CQ/amine_high', 'LUB-CQ/amine_low', 'LUB-TPO_high' and 'LUB-TPO_low', with a photoinitiator concentration of, respectively, 2.0 wt% CQ and 2.0 wt% EDMAB (amine), 0.35 wt% CQ and 0.35 wt% amine, 2.0 wt% TPO, and 0.35 wt% TPO, were used in this study (Table 1). The experimental LUB adhesives were provided by Kuraray Noritake without photoinitiator. The photoinitiators CQ in combination with the co-initiator EDMAB and TPO were purchased from Sigma-Aldrich (Sigma-Aldrich Chemie, Steinheim, Germany). The respective amounts of CQ and EDMAB, and of TPO were measured on an analytical balance with a 0.01-mg accuracy (AB304-S' analytic balance; Mettler-Toledo, Greifensee, Switzerland); the photoinitiator was added to the LUB adhesive in light-shielding amber vials that were extra wrapped with aluminum foil to shield protect the solution from light. The photoinitiator was dissolved in the adhesive using a closed container that was immersed in an ultrasonic bath (Bandelin Sonorex; Bandelin Electronic, Berlin, Germany) for 1 min and then homogeneously mixed using a rotating machine (Rotator; Agar Scientific, Essex, United Kingdom) for 24 h, this following the recipe described in detail in the previous study [4].

2.2. Mini-iFT

Fifty non-carious human third molars (collected following informed consent approved by the Commission for Medical Ethics of KU Leuven under the file number S57622), stored in 0.5% chloramine T/water at 4 °C, were used within three months after extraction. The occlusal third of the crown was removed with a diamond saw (Isomet 1000; Buehler, Lake Bluff, IL, USA), exposing a flat mid-coronal dentin surface, which was wet-sanded with 320-grit SiC paper (Buehler-Met II SiC wet grinding paper; Buehler, Lake Bluff, IL, USA) to produce a standard smear layer resembling that produced by a regular diamond bur. All dentin surfaces were carefully examined for remaining enamel and pulp tissue using a stereo-microscope (Stemi 2000 CS; Carl Zeiss, Jena, Germany). Each adhesive was applied following the instructions of Kuraray Noritake for the commercial adhesive Clearfil S^3 Bond Plus by actively rubbing the adhesive onto the dentin surface for 10 s, followed by 5 s gentle air-drying until the adhesive no longer moved. The adhesive formulations were next light-cured for 10 s using a polywave LED light-curing unit (Bluephase 20i; Ivoclar Vivadent, Schaan, Liechtenstein), employed in 'high mode', with an output of around 1100 mW/cm^2; the radiant exposure was 2.9 J/cm^2 up to 420 nm and 14 J/cm^2 above 420 nm, as was measured by a MARC Resin Calibrator (BlueLight Analytics, Halifax, NS, Canada). A composite build-up (Filtek Z100; 3 M ESPE, Seefeld, Germany: shade A2, lot N459523) was made in layers using a polytetrafluoroethylene mold (8 × 8 × 10 mm). The root of the tooth was removed 3 mm below the adhesive-dentin interface and a similar composite build-up was made at the root side using the self-etch adhesive Clearfil SE Bond (Kuraray Noritake). After 1 week water storage at 37 °C, the specimens were sectioned perpendicular to the interface using a semi-automatic high-speed diamond saw (Accutom-50; Struers, Ballerup, Denmark: feed speed of 0.075 mm/s, wheel speed at 4000 rpm) with a water-cooled diamond blade with a diameter of 102 mm and a thickness of 300 μm (M1D10; Struers) to obtain rectangular sticks (micro-specimens of 1.5 × 2.0 mm wide and 16–18 mm