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Aspects of enamel bonding using experimental silanes for orthodontic adhesion

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ABSTRACT

This *in vitro* study investigates bonding to enamel using experimental silane-based primers with and without 2-hydroxyethylmethacrylate (HEMA) under various artificial ageing methods. One hundred and fifty sound extracted human premolars were used and randomly assigned to three experimental study groups. They were first acid-etched for 15 s, rinsed with water spray, air dried, and applied 0.3 ml of artificial saliva on the enamel surfaces. Two groups of enamel surfaces were primed using silane-based experimental primers (1.0 vol% of 3-isocyanatopropyltrimethoxysilane and 0.5 vol% of *bis*-1,2-(triethoxysilylethane) with and without 25% HEMA) while one group was served as control. Then, stainless steel premolar orthodontic brackets were fixed onto teeth with orthodontic resin composite. The specimens from each group ($n=10$) were stored under different ageing conditions: thermo-cycling (500, 2000, and 6000 cycles), storage in artificial saliva for 24 h, and for one year. The shear adhesion (bond) strength (SBS) was tested by using a universal testing machine at a crosshead speed of 1.0 mm/min. Surface morphology and failure modes at the debonded interfaces were examined using SEM. Two-way ANOVA and *post hoc* tests were used to compare the SBS ($\alpha=0.05$). The results suggested that an experimental primer with 25% HEMA, after 24 h storage in artificial saliva, produced the highest mean SBS (22.1 MPa, SD 2.2 MPa). The lowest mean value (5.8 MPa, SD 1.1 MPa) was obtained with the control group thermo-cycled (6000 cycles). There was a significant difference between the experimental primers ($p<0.001$) and artificial ageing ($p<0.001$). We conclude that 25% HEMA inclusion in silane primer could provide satisfactory adhesion strength, and 500 cycles of thermo-cycling (ISO TR 11450) does not correlate with 1-year artificial saliva storage for enamel bonding test.

1. Introduction

In orthodontics, various methods are used to increase adhesion of the orthodontic brackets to enamel surfaces [1–3]. One of these methods is the application of “primers” to enamel surface to promote bonding [4,5]. “A primer” *per se* in dentistry could be broadly defined as a pretreatment liquid that can improve the surface wetting to allow a stronger adhesive bond between the applied surface and an adhesive, such as a resin composite luting cement. The extensive use of “a restorative primer” and "an orthodontic primer” is a successful method to promote adhesion of restorative materials [6,7] and in orthodontics [8]. One of the commonly used ingredients in commercial dental primers is 2-hydroxyethylmethacrylate (HEMA), which has a pendant and hydrophilic hydroxyl (− OH) group at one end and a polymerizable hydrophobic $>C=C<$ group at the other end [7]. A study had shown the incorporation of HEMA into the formula of a silane-based restorative primer could increase the bond strength of restorative resins to tooth substrates as a result of adhesion of hydrophobic resin-composites (filled resins) to hydrophilic dentine and enamel [9]. It seems to be silanes being as hybrid inorganic–organic functional monomers could be used as potential monomers for adhesion promoters in orthodontics.

The use of acid etching combined with adhesion promoters (primers) is required when orthodontic brackets are cemented to enamel using a resin-based orthodontic adhesive [10,11]. Despite the incorporation of 10–25 vol% HEMA in dental primers was proved to increase the bond strength of orthodontic adhesives to enamel surfaces, the incorporation of HEMA in larger quantities in adhesive resins could decrease the bond strength, possibly due to HEMA could attract water into the resin matrix as this may initiate detachment of the inorganic fillers which lead to degradation of the adhesive material [12,13]. A study has suggested that the

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addition of a hydrophilic HEMA to a silane-based primer could result in better encapsulation of the crystallite in the outer enamel, enhancing durable bonding [14]. Another study stated that applying a hydrophilic component to a polar solvent, such as ethanol or water, could adversely affect the bond strength to enamel [15]. Furthermore, there is an adverse proportionality between the amounts of the applied primer and the mechanical properties. Application of high amounts of a restorative primer could produce less values of bond strength when compared to less amounts of primer. This could be attributed to incomplete evaporation of the solvent from the primer solution [16]. Thus, some HEMA-free adhesives have been developed to minimise the effect of water sorption and hence degradation of the adhesive [17]. However, one study [13] contrasted and compared various solvents to challenge the bonding system but it was deemed not to be as clinically relevant as an artificial ageing. Furthermore, such solvents could be selected on purpose, not randomly. Therefore, to substantiate a fair comparison to test the bonding systems, a careful selection of test methods is vital.

Several studies have employed various techniques to study the effect of durability of some orthodontic adhesives and orthodontic primers. It is unclear whether such durability is an intrinsic or an external factor, such as the applied load [5,8,18]. In fact, clinically, fixed orthodontic appliances are subjected to repeated thermal and mechanical stresses inside the oral cavity. Artificial ageing by thermo-cycling or storage in artificial saliva, for long periods is a common *in vitro* method to predict the effect of these stresses on the long-term use of orthodontic appliances. Various studies has shown immersion of teeth in liquids, *e.g.* distilled water or artificial saliva, for long periods of time is one of the methods that can be used to test the durability of the orthodontic adhesives [19–21], despite liquid storage tests are time consuming and need tedious follow-up. In addition to the storage in artificial saliva, one of the reliable techniques to simulate intraoral temperature is the artificial ageing of the tooth substrates by using the so-called thermo-cycling process. It mimics the changes in temperature that may occur intra-orally [22,23]. According to the ISO TR 11450 [24], a protocol of thermo-cycling with 500 cycles in water between 5 and 55 °C, is sufficient to estimate one year of ageing. However, other studies have concluded that a higher number of cycles should be performed to estimate the one-year ageing. Some of these protocols have suggested different thermo-cycles such as 500, 1000, 2000 [25–27] and up to 10000 cycles [28]. At present, there is no general consensus about the cycles to properly simulate the one-year ageing. Nevertheless, due to the vast variety of chemicals in the bonding systems, the number of cycles should be particularly specified for each bonding system. For example, the relevant cycles could be classified according to the chemical consistency of the primer. Thereby, a parallel-run comparison, *i.e.,* in real time and by thermo-cycling, would be necessary to validate the specificity of such a system, as well as to better understand the nature of material degradation at the interfaces between enamel, adhesives and brackets for various orthodontics bonding systems.

The null hypotheses tested were: (1) a HEMA addition in silane-based primers has no effect on adhesion bond strength of orthodontic brackets, regardless of the ageing method, and: (2) increasing the number of cycles in thermo-cycling has no influence on durability of the shear bond strength.

2. Materials and methods

2.1. Selection of teeth

One hundred and fifty sound human premolar teeth extracted for orthodontic purposes were selected for this *in vitro* study. Teeth selection criteria were based on absence of restorations, caries and damage free, and no history of chemical treatment, *e.g.,* bleaching. Immediately after extraction, all the teeth were kept in distilled water containing 0.2% (w/v) thymol to inhibit bacterial growth [29,30] for one day. The teeth were then embedded into acrylic moulds for the next steps.

2.2. Preparation of experimental primers

The silane monomers used in the experimental primers, 3-isocyanatopropyltrimethoxysilane (1.0 vol%, by Gelest, Morristown, PA, USA), and *bis*-1,2-(triethoxysilylethane), were diluted in 95% ethanol, with an addition of 1 M acetic acid, as described in the literature [31,32]. Finally, an amount corresponding to 25% HEMA was added in one of the primers.

2.3. Experimental design

The specimens were randomly assigned to one of the three experimental groups of 50 each. All the specimens were acid-etched with a commercially available etching gel (37.5% phosphoric acid, Gel-etchant 3, SDS Kerr, USA) for 15 s, and then rinsed with water spray and air-dried. This was to simulate the clinical operation. Next, 0.3 ml of artificial saliva was applied as a contaminant to the enamel surface area of the teeth by using a fine brush (Kerr, USA). The artificial saliva buffer was prepared (Biochemistry Unit, Faculty of Dentistry; UIA-Malaysia) and modified from the SAGF medium formula [33] to cope with the protocol of the study. The enamel surfaces were treated as follows:

Group I ($n=50$): a HEMA-free experimental silane-based dental primer was applied.

Group II ($n=50$): an experimental silane-based dental primer with an addition of 25 vol% HEMA was applied.

Group III ($n=50$): No application of primer solution (Non-primed specimens were considered as controls).

Stainless steel premolar orthodontic brackets with mesh bases were used (Gemini, 3M Unitek, USA). An average surface area of the bracket bases ($\sim$13.0 mm^2), was measured by using a digital caliper (Absolute, Mitutoyo, Kawasaki, Japan). The brackets were fixed in place by using an orthodontic self-adhesive resin (One-Step Ortho-Adhesive, Alpha Dent, USA). Each bracket was exposed to a compressive force of 3 N (a constant load of 300.0 g) for 10 s using CORREX Force Gram Indicator with a round blunt feeler 48 mm (Correx Co, Bern, Switzerland).

The following artificial ageing procedures were performed for each group:

Subgroup A ($n=10$): one-year storage in freshly prepared artificial saliva. The specimens were kept immersed in freshly prepared artificial saliva at 37 °C, after addition of new artificial saliva in 30-day intervals, for one year.

Subgroup B ($n=10$): Thermo-cycling for 500 cycles in water baths, between 5 and 55 °C, according to the (ISO) TR 11450 standards (1994).

Subgroup C ($n=10$): Thermo-cycling for 2000 cycles in water baths between 5 and 55 °C. Subgroup D ($n=10$): Thermo-cycling for 6000 cycles in water baths between 5 and 55 °C. Subgroup E ($n=10$): Initial SBS after 24 h storage in freshly prepared artificial saliva.

2.4. Shear adhesion strength test

Shear adhesion (bond) strength (SBS) was determined by debonding the brackets with a vertically directed moving knife arm. A precision universal testing machine (AGS-X, Shimadzu, Japan) at a crosshead speed of 1.0 mm/min was used.