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The role of HEMA in one-step self-etch adhesives

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ABSTRACT

In spite of its high allergenic potential, 2-hydroxyethyl methacrylate (HEMA), a low-molecular-weight monomer, is frequently used in adhesives for its positive influence on the bond strength. In addition, the presence of HEMA in one-component one-step adhesives can prevent phase separation.

Objectives. In search of improved bonding effectiveness, the 24-h bond strength of four experimental one-step self-etch adhesives with different concentrations of HEMA to bur-cut enamel and dentin was determined using a micro-tensile bond strength protocol.

Methods. The tested experimental adhesives (Exp-0, Exp-10, Exp-19 and Exp-36) only differed in their concentration of HEMA, which was 0, 10, 19 and 36%, respectively. With an increasing concentration of HEMA, the concentration of acetone was decreased. Besides bond strength, the adhesives were also examined by light-microscopy for phase separation. The interface was investigated by SEM and TEM.

Results. Regarding bond strength, Exp-10 performed best. Even though Exp-36 was the only adhesive formulation that did not exhibit phase separation on a glass plate, it yielded the lowest bond strength. Accordingly, droplets could be observed by SEM and TEM in the adhesive layers of all adhesives, except for Exp-36 on enamel.

Conclusion. A small amount of HEMA (10%) improved the bond strength of a one-step self-etch adhesive. When added in higher concentrations, this beneficial effect of HEMA on the bond strength is lost due to increased osmosis, which resulted in many droplets; due to reduced polymerization conversion; and sub-optimal physico-mechanical properties of the resultant poly-HEMA containing adhesive interface.

1. Introduction

2-Hydroxyethyl methacrylate (HEMA) is a water-soluble methacrylate monomer frequently present in dental adhesives [1]. Its polar properties and small dimensions enhance the wetting properties of the adhesive solution [2,3] and the penetration efficacy of the adhesive into demineralized dentin [4]. HEMA has been reported to positively influence bond strength to dentin [5]. Because of its hydrophilic character, it is also frequently added to improve miscibility of hydrophobic and hydrophilic components in an adhesive solution [6–8].

The major drawback of the use of HEMA is its high allergenic potential. Fast penetration of this monomer through gloves and through skin due to its low molecular weight can

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cause contact dermatitis (allergic reaction type IV), which is a well-known occupational disease among dentists [9–11]. As HEMA is not exclusively used in dentistry (varnish, paints, lenses, fake nails, etc.), cross-allergic reactions have also been described [11]. Beside that, HEMA, being a mono-methacrylate, may also deteriorate the mechanical properties of the polymerized adhesive, which can result in inferior mechanical strength of the cured adhesive, enhanced water-uptake, swelling and staining [8,12].

Recently, HEMA was shown to play an important role in preventing phase-separation reactions in one-component one-step self-etch adhesives [6]. In these 'one-bottle' solutions, polar and apolar ingredients are blended together with a solvent such as water, acetone and ethanol. The solvent keeps the ingredients in solution, but once dispensed subsequent evaporation of the solvent can trigger a phase-separation reaction with the formation of multiple droplets. Macroscopically, this can be observed as an uncured drop of adhesive solution that becomes opaque. Microscopically, many droplets in the uncured adhesive can be seen. When the adhesive is cured before the end of the separation reaction and thus before complete removal of the droplets, the droplets get frozen in the adhesive layer. HEMA can prevent such a phase-separation reaction by replacing the solvent and keeping the ingredients in solution [1].

The objective of this study was first to investigate the effect of HEMA on the adhesive performance of one-component one-step self-etch adhesives and second to determine the concentration of HEMA needed to prevent phase separation. The null hypothesis advanced in this study was that HEMA did not influence the bond strength of one-step self-etch adhesives.

2. Materials and methods

Four experimental mild (pH 2) one-component (one-bottle) one-step self-etch adhesives containing a carboxylate and phosphate-based functional monomer were prepared. Their ingredients are listed in Table 1. They had an identical composition but for the ingredient HEMA. Exp-0, Exp-10, Exp-19 and Exp-36 contained respectively 0, 10, 19 and 36% of HEMA. With an increasing concentration of HEMA, the concentration of the solvent acetone was reduced. In Exp-36 the concentration of acetone was substantially reduced.

3. Light-microscopy

All adhesive solutions were examined (uncured) for a potential phase-separation reaction and the presence of droplets by light-microscopy (LM) (Olympus BH2, Hamburg, Germany). A drop of each self-etching solution was dispensed onto a glass plate, and imaged real-time at different magnifications (140–280×) using a digital camera (JVC TK-870E, Yokohama, Japan).

4. μTBS-testing

The bond strength to enamel and dentin was determined using a standardized micro-tensile bond strength protocol. Non-carious human third molars (gathered following informed consent approved by the Commission for Medical Ethics of KULeuven) were stored in 0.5% chloramine/water at 4 °C and used within 1 month after extraction. To prepare dentin samples, the occlusal crown third was removed with a diamond saw (Isomet 1000, Buehler, Lake Bluff, IL, USA), thereby exposing a flat mid-coronal dentin surface. A standardized bur-cut smear layer was produced by removing a thin layer of the surface using a Micro-Specimen Former (University of Iowa, Iowa City, IA, USA), equipped with a high-speed regular-grit (100 μm) diamond bur (842, Komet, Lemgo, Germany). For enamel, a flat surface was ground using the same bur at the buccal and lingual surface of a tooth. All four adhesives were applied on air-dried, but not overly desiccated enamel and dentin. After a dwell time of 10 s, the adhesive was strongly air-blown with maximum pressure and subsequently light-cured for 10 s (Table 1). Composite build-ups were made with Gradia Direct Anterior (GC, color A2) in three or four layers to a height of 5–6 mm. After 24 h storage in distilled water (37 °C), rectangular sticks (2 mm × 2 mm wide; 8–9 mm long) were sectioned perpendicular to the adhesive-tooth interface using the Isomet saw. Only the four central dentin sticks were used to eliminate substrate regional variability [13,14]. The sticks were trimmed at the interface into an hourglass shape (diameter of ±1.1 mm) using the Micro-Specimen Former, equipped with a fine-grit (30 μm) diamond bur (5835KREF, Komet) in a high-speed handpiece under air/water coolant. The specimens were fixed to Ciucchi's jig with cyanoacrylate

Table 1 – Adhesives investigated, their composition and application procedure

Adhesive	Manufacturer	Composition[a,b]	Application
Exp-0	GC, Tokyo, Japan	4-MET, phA-m, DMA, acetone, water, filler, photoinitiator, stabilizer	(1) Apply adhesive to the entire surface with a disposable applicator
Exp-10		HEMA (10%), 4-MET, phA-m, DMA, acetone, water, filler, photoinitiator, stabilizer	(2) Keep dentin wet (shiny surface) with adhesive for at least 10 s; strongly air-dry
Exp-19		HEMA (19%), 4-MET, phA-m, DMA, acetone, water, filler, photoinitiator, stabilizer	
Exp-36		HEMA (36%), 4-MET, phA-m, DMA, acetone, water, filler, photoinitiator, stabilizer	(3) Light-cure for at least 10 s

[a] Abbreviations of monomers in alphabetical order: DMA, dimathacrylates; HEMA, 2-hydroxyethyl methacrylate; 4-MET, 4-methacryloxyethyltrimellitic acid; phA-m, phosphoric acid ester monomer.
[b] Underlined words refer to differences in composition.