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HEMA release and degree of conversion from a resin-modified glass ionomer cement after various delays of light activation

Elisabeth Dursun[a,b,*], *Jean-François Nguyen*[c,d], *Mie-Leng Tang*[a], *Jean-Pierre Attal*[a,e], *Michael Sadoun*[a]

[a] *Research Unit in Dental Material, Innovations and Interfaces, EA4462, Faculty of Dental Surgery, Paris Descartes University, 1 rue Maurice Arnoux, 92120 Montrouge, France*
[b] *Albert Chenevier Hospital, 40 rue de Mesly, 94000 Créteil, France*
[c] *UFR d'Odontologie, Université Paris Diderot, Paris, France*
[d] *PSL Research University, Chimie ParisTech—CNRS, Institut de Recherche de Chimie Paris, 75005 Paris, France*
[e] *Charles Foix Hospital, 7 avenue de la République, 94200 Ivry-sur-Seine, France*

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ABSTRACT

Objective. The setting of resin-modified glass ionomer cements (RMGIC) involves the acid–base reaction and the polymerization of HEMA monomers. Each setting mechanism may compete with the other. The aim of this study was to determine an optimum polymerization after various delays of light-activation, to minimize the release of free HEMA and to better understand the setting mechanism of RMGICs. The null hypothesis tested was: increasing the delay before light-activation of an RMGIC has no effect on HEMA release and on its degree of conversion (DC).

Methods. Five groups were investigated: (a) control group with no light-curing; (b) light-curing delayed 1 min after mixing; (c) delayed 5 min; (d) delayed 10 min; (e) delayed 15 min. For each, HEMA release was analyzed with high performance liquid chromatography at 1 h, 6 h, 24 h, 7 days, 14 days and 28 days and the DC was tested by ATR-FTIR spectrometer. Data were analysed by one-way ANOVA, followed by Scheffe multiple mean comparisons.

Results. A delay in light-activation caused a significant increase in the cumulative HEMA release. The highest release was determined for the group without light-curing. The DC increased significantly when the delay was increased, until a maximum value for 10 min delay, then decreased when the delay was more extended, with a minimum value for the group without light-curing.

Significance. A short delay before light-curing could limit the HEMA release and could be more biocompatible. The results highlighted the competition between the acid–base and the polymerization reactions. They also showed there is no correlation between the monomer release and the DC.

* *Corresponding author at*: Research Unit in Dental Material, Innovations and Interfaces, EA4462, Faculty of Dental Surgery, Paris Descartes University, 1 rue Maurice Arnoux, 92120 Montrouge, France. Tel.: +33 1 58 07 67 25.
E-mail address: elisabethdursun@gmail.com (E. Dursun).
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1. Introduction

In order to improve the glass ionomer cements' properties, it was proposed to incorporate resin components, generally monomers of HEMA. These resin-modified glass ionomer cements (RMGICs) offer greater mechanical strength [1], increased dental bond strength [2], decreased early moisture sensitivity [3] and extended working time.

However, the polymerization of light-cured resin materials is always incomplete. The degree of conversion (DC), as defined as the percentage of double bonds reduced during the polymerization process, was determined in the 33–75% for RMGICs after 24 h [4,5]. Consequently, the leaching of unreacted monomers causes increasing solubility in the oral environment and, in fine, microleakage.

The presence of free HEMA, released from RMGICs [6–8], and its diffusion at the restorative/dentin interface after completion of polymerization [9] were reported. The leached residual monomers were shown to easily diffuse through the dentinal tubules due to their hydrophilic property and low molecular weight, reaching dental pulp cells. All these phenomena may result in recurrent decays and/or irritation of the pulp [10,11].

Conjointly, the addition of HEMA [12] and/or photopolymerization [5] would significantly reduce the ionic reaction rate. The setting of RMGICs involves two reactions: an acid–base reaction between polyalkenoic acid and fluoro-alumino-silicate glass particles, and the polymerization of HEMA monomers. These reactions may rely on and/or compete one another [13,14]. Time of light-activation after mixing could determine the ratio between both reactions. No previous studies have evaluated the HEMA release of RMGICs after various delays in light-activation and its possible correlation with the degree of conversion.

The aim of this study was to determine optimum polymerization conditions by varying the time between mixing and light-activation. It was postulated that the aforementioned time delay may help minimize the release of free HEMA and resolve the setting mechanism of RMGICs. The null hypothesis tested was as follows: increasing the delay before light-activation of an RMGIC has no effect on HEMA release and its degree of conversion (DC).

Analysis of HEMA release (absolute and relative rates) from the RMGIC test sample as a function of the delay time between sample mixing and light-activation was carried out by high performance liquid chromatography (HPLC); in each case, HEMA degree of conversion was determined by attenuated total reflectance – Fourier transform Infrared (ATR-FTIR) spectrophotometry.

2. Materials and methods

2.1. Material and experimental conditions

The RMGIC selected for this study was the Fuji II^{TM} LC (GC) supplied in capsules. The manufacturer product specifications including batch number and composition, sample set designation and polymerization conditions are summarized in Table 1.

The experimental design consisted in 5 sample sets spanning various light-curing delays *i.e.*, no light-curing (control NP), light-curing 1 min after mixing (P1), 5 min after mixing (P5), 10 min after mixing (P10) and 15 min after mixing (P15) respectively.

2.2. Analysis of HEMA release by HPLC

2.2.1. Sample preparation

A sample set was defined as nine replicate bars, which were prepared in ($25 \times 2 \times 2$)mm silicone molds. Each capsule was mixed 10 s in a mechanical mixer and injected into the mold. The mold was covered with a Mylar strip; a glass plate was placed atop the Mylar strip to help apply even pressure on the sample and express excess material, resulting in a homogeneous flat surface. The bars were then light-cured with a series of three 20 s irradiations using a LED unit (*Radii, SDI, Victoria, Australia*) set to a light output power of 1200 mW/cm^2 as measured with a commercial radiometer (*Dentsply Caulk, Milford, USA*). Irradiation was applied in the middle and at each extremity of each given sample. Surface area and weight were measured and recorded.

All the samples were kept in the dark before the initiation of the photopolymerization and then prepared under dimmed conditions to reduce background reaction due to ambient light [15].

2.2.2. HPLC analysis

The HPLC analysis was conducted using an Agilent 1260 Infinity Quaternary LC (*Agilent Technologies, Waldbronn, Germany*), equipped with a quaternary pump (model G1311B), a temperature-controlled column unit. Detection was performed by UV-diode array (model G4212B). Separation was achieved at room temperature using a Poroshell 120 EC-C18 column (4.6×50 *mm, 2.7* μ*m, Agilent Poroshell, USA*) in isocratic conditions with a 60% (v) solution of HPLC grade acetonitrile in HPLC grade water (*Fisher Scientific, Bishop Meadow Road, UK*) at a flow rate of 1 mL/min and a run time of 4 min. The HEMA retention time in the aforementioned run conditions was determined at 0.61 min, using HEMA peaks registered for standard solutions. A complete UV spectrum was acquired for each sample; quantification was possible by analysis of the HEMA peak area at the 214 nm wavelength.

For each sample set, the bars were tested by HPLC for HEMA release as groupings of three (*i.e.*, triplicate per set). Three bars were placed in a 20 mL glass vial and 10 mL of a 75% (v) solution of ethanol in deionized water (HPLC grade ethanol and grade water, *Fisher Scientific, Bishop Meadow Road, UK*) were added. The vials were sealed and placed in a 37 °C incubator (*Memmert, Schwabach, Germany*). A series of three 20 μL aliquots from each vial was taken with a microsyringe at the following storage time points, 1 h, 6 h, 24 h, 7 days, 14 days and 28 days, and injected onto the instrument for analysis.

For quantitative analysis, a calibration curve was constructed using four 10 mL solutions of HEMA (1×10^{-3}; 1×10^{-4}; 1×10^{-5}; 1×10^{-6})M prepared by serial dilution of a 1×10^{-3} M HEMA stock (in 75% (v) ethanol). The resulting standard solutions were stored at ambient temperature. Linearity of the calibration curve, based on the quantitative determination of HEMA in the four solutions, was assessed by