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Effect of layer thickness on the elution of bulk-fill composite components

Lena Rothmund[a,b], *Franz-Xaver Reichl*[a,b], *Reinhard Hickel*[a], *Panorea Styllou*[a,b], *Marianthi Styllou*[a,b], *Kai Kehe*[b,c], *Yang Yang*[a,b], *Christof Högg*[a,b,*]

[a] *Department of Operative/Restorative Dentistry, Periodontology and Pedodontics, Ludwig-Maximilians-University of Munich, Goethestr. 70, 80336 Munich, Germany*

[b] *Walther-Straub-Institute of Pharmacology and Toxicology, Nussbaumstr. 26, 80336 Munich, Germany*

[c] *Bundeswehr Institute of Pharmacology and Toxicology, Neuherbergstr. 11, 80937 Munich, Germany*

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ABSTRACT

Objective. An increment layering technique in a thickness of 2 mm or less has been the standard to sufficiently convert (co)monomers. Bulk fill resin composites were developed to accelerate the restoration process by enabling up to 4 mm thick increments to be cured in a single step. The aim of the present study is to investigate the effect of layer thickness on the elution of components from bulk fill composites.

Methods. The composites ELS Bulk fill, SDR Bulk fill and Venus Bulkfill were polymerized according to the instruction of the manufacturers. For each composite three groups with four samples each ($n=4$) were prepared: (1) samples with a layer thickness of 2 mm; (2) samples with a layer thickness of 4 mm and (3) samples with a layer thickness of 6 mm. The samples were eluted in methanol and water for 24 h and 7 d. The eluates were analyzed by gas chromatography/mass spectrometry (GC/MS).

Results. A total of 11 different elutable substances have been identified from the investigated composites. Following methacrylates showed an increase of elution at a higher layer thickness: TEGDMA (SDR Bulk fill, Venus Bulk fill), EGDMA (Venus Bulk fill). There was no significant difference in the elution of HEMA regarding the layer thickness. The highest concentration of TEGDMA was 146 μg/mL for SDR Bulk fill at a layer thickness of 6 mm after 7 d in water. The highest HEMA concentration measured at 108 μg/mL was detected in the methanol eluate of Venus Bulk fill after 7 d with a layer thickness of 6 mm.

Significance. A layer thickness of 4 mm or more can lead to an increased elution of some bulk fill components, compared to the elution at a layer thickness of 2 mm.

* *Corresponding author at*: Department of Operative/Restorative Dentistry, Periodontology and Pedodontics, Ludwig-Maximilians-University of Munich, Goethestr. 70, 80336 Munich, Germany. Tel.: +49 89 2180 73809; fax: +49 89 2180 73817.
E-mail address: christof.hoegg@lrz.uni-muenchen.de (C. Högg).

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1. Introduction

In the last decade the use of resin based composites (RBCs) has increased tremendously. RBCs, consisting of a number of (co)monomers and additives, belong to the most commonly used filling materials. Due to the incomplete (co)monomer-polymer conversion, a release of the unpolymerized (co)monomers from the dental composite is described [1,2]. There are many *in vitro* studies on the toxicity and biocompatibility, which have shown that some of the eluted (co)monomers and additives even have estrogenic, mutagenic, teratogenic and genotoxic effects [3–6]. Previous *in vivo* studies have demonstrated that HEMA, TEGDMA and BisGMA can be metabolized to the expoxy compound 2,3-epoxymethacrylic acid in hepatic microsomes [7–9]. Epoxides are regarded as mutagenic and carcinogenic agents [10–12].

The final degree of conversion (DC) depends mainly on intrinsic factors such as the chemical structure of the (co)monomer and photo initiator concentration and extrinsic factors such as polymerization conditions and curing modes [13,14]. The energy of the light emitted from a light curing unit decreases drastically when transmitted through a rinsing composite [15]. Thus far, an increment layering technique in a thickness of 2 mm or less has been the standard to sufficiently convert (co)monomers [16].

A new category of RBCs, bulk-fill resin composites, has been introduced over the past few years. They were developed to accelerate the restoration process by enabling up to 4 mm thick increments to be cured in a single step, thereby skipping the time-consuming layering process. The manufacturers explain that the higher depth of cure of the bulk-fill resin composites is due to the more potent initiator system or/and higher translucency. Studies have already been performed on the mechanical properties of bulk-fill composites [17–22]. Thus, for example, for cuspal deflection [22], the marginal integrity of a filling [20,21], just as for its cure depth [21] better results of bulk-fill composites, compared to composites which are added in the incremental technique were detected. However, also adverse results were found compared to conventional composites such as the conversion rate, for bulk-fill composites [23]. A conversion rate >55% for bulk-fill composites is still in the clinically acceptable range but it is still less than for conventional composites [23].

It was shown that the elution of bulk-fill composites is comparable to that of conventional materials despite their increased layer thickness of 4 mm [24] and amount of eluted (co)monomers increases with elution time [25,26].

However, there are no data available to what extent a layer thickness of up to 6 mm, in comparison to a layer thickness of 2 and 4 mm, has an effect on the amount of elutable components from bulk fill composites. The aim of the present study is therefore to clarify the effect of layer thickness on the elution of components from bulk fill composites. In the null hypothesis it is assumed that a variation of layer thickness does not have an influence on the concentration of eluted substances from bulk fill composites.

2. Materials and methods

The tested composites including manufacturers' data are listed in Table 1.

2.1. Preparation of samples

Composites (Table 1) were polymerized exactly according to instruction of the manufacturer. For each composite three groups with four samples each ($n = 4$) were prepared: (1) samples with a layer thickness of 2 mm; (2) samples with a layer thickness of 4 mm and (3) samples with a layer thickness of 6 mm. For the preparation of the samples, polytetrafluoroethylene (PTFE) rings with a diameter of 6 mm were used. The PTFE rings were filled with uncured dental material, covered with plastic strips (Frasaco, Tettnang, Germany) to prevent the formation of an oxygen inhibition layer and were finally polymerized with a LED-lamp (Elipar S^{TM}10® high intensity halogen light, 1200 mW/cm^2, 3 M ESPE, Seefeld, Germany) in accordance with the manufacturer's instructions (Table 1). The curing unit was directly applied on the sample's surface. The light intensity of the LED-lamp was controlled with Demetron® Radiometer (Kerr, USA) and was always between 1100 and 1200 mW/cm^2. Samples had approximately a volume of 56.6, 113.1 and 169.7 mm^3, and surface area of 94.3, 132.0 and 169.7 mm^2 at a layer thickness of 2, 4 and 6 mm, respectively.

Subsequently, samples were incubated (face up) in brown glass vails (Macherey-Nagel, Düren, Germany) with 1 ml of methanol (GC Ultra Grade, RATISOLV® ≥99.9%, Roth, Karlsruhe, Germany) or 1 ml water (LC-MS-Grade, ROTISOLV®, Roth, Karlsruhe, Germany) and stored in the dark at 37 °C and analyzed after 1 d and 7 d by gas chromatography/mass spectrometry (GC/MS) [27]. 100 μl of the water eluates were previously extracted one time with 100 μl ethyl acetate (LC-MS-Grade, ROTISOLV® ≥99.9%, Roth, Karlsruhe, Germany) (1:1 v/v). To optimize layer separation, the samples were centrifuged at 2800 rpm for 10 min [28].

As internal standard caffeine (CF) solution (0.01 mg/ml) (HPLC ≥99.0%, Sigma Aldrich, St. Louis, United States) was added.

2.2. Analytical procedure

The analysis of the eluates was performed on a Finnigan Trace GC ultra gas chromatograph connected to a DSQ mass spectrometer (Thermo Electron, Dreieich, Germany). A J&W VF-5ms capillary column (length 30 m, inner diameter 0.25 mm; coating 0.25 μm; Agilent, Böblingen, Germany) was used as the capillary column for gas chromatographic separation. Helium 5.0 was used as carrier gas at a constant flow rate of 1 ml/min. The temperature of the transfer line was 250 °C. For sample analysis 1 μL each was injected in splitless mode (splitless time 1 min, split flow 50 ml/min). For capillary transfer the programmable temperature vaporizing (PTV) inlet was heated from 30 °C to 320 °C (14.5 °C/s) and finally held for five min at this temperature. The GC oven was initially heated isothermally at 50 °C for 2 min, then increased to 280 °C (25 °C/min) and finally remained for five min at this temperature. The mass spectrometer (MS) was operated