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Elution of monomer from different bulk fill dental composite resins

Mehmet Ata Cebe[a], Fatma Cebe[a,*], Mehmet Fatih Cengiz[b], Ali Rıza Cetin[c], Osman Fatih Arpag[d], Bora Ozturk[c]

[a] Abant Izzet Baysal University, Faculty of Dentistry, Department of Restorative Dentistry, Bolu, Turkey
[b] Akdeniz University, Food Safety and Agricultural Research Center, Antalya, Turkey
[c] Selcuk University, Faculty of Dentistry, Department of Restorative Dentistry, Konya, Turkey
[d] Mustafa Kemal University, Faculty of Dentistry, Department of Periodontology, Hatay, Turkey

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ABSTRACT

Objective. The purpose of this study was to evaluate the elution of Bis-GMA, TEGDMA, HEMA, and Bis-EMA monomers from six bulk fill composite resins over four different time periods, using HPLC.

Methods. Six different composite resin materials were used in the present study: Tetric Evo Ceram Bulk Fill (Ivoclar Vivadent, Amherst, NY), X-tra Fill (VOCO, Cuxhaven, Germany), Sonic Fill (Kerr, Orange, CA, USA), Filtek Bulk Fill (3M ESPE Dental Product, St. Paul, MN), SDR (Dentsply, Konstanz, Germany), EQUIA (GC America INC, Alsip, IL). The samples (4 mm thickness, 5 mm diameter) were prepared and polymerized for 20 s with a light emitted diode unit. After fabrication, each sample was immediately immersed in 75 wt% ethanol/water solution used as extraction fluid and stored in the amber colored bottles at room temperature. Ethanol/water samples were taken (0.5 mL) at predefined time intervals:10 m (T1), 1 h (T2), 24 h (T3) and 30 days (T4). These samples were analyzed by HPLC. The obtained data were analyzed with one-way ANOVA and Tukey HSD at significance level of $p < 0.05$.

Results. Amount of eluted Bis-EMA and Bis-GMA from Tetric Evo Ceram Bulk Fill and amount of eluted TEGDMA and HEMA from X-tra Fill higher than others composites ($p < 0.05$).

Significance. Residual monomers were eluted from bulk fill composite resins in all time periods and the amount of eluted monomers was increased with time.

1. Introduction

Dental composite resin materials consist of a resin matrix, inorganic filler, and a coupling agent. The common monomers used in the resin matrix are bisphenyl-glycidyl-methacrylate (Bis-GMA), triethylene glycol dimethacrylate (TEGDMA), urethane dimethacrylate (UDMA), ethoxylated bisphenol A dimethacrylate (Bis-EMA) [1].

The degree of conversion (DC) of a resin composite is crucial in determining its biocompatibility [2]. It has been shown that decreases in DC might lead to a decrease in the

* *Corresponding author*. Tel.: +90 05073191050.
E-mail addresses: atacebe014@hotmail.com (M.A. Cebe), fkahveci61@hotmail.com (F. Cebe), fcengiz@akdeniz.edu.tr (M.F. Cengiz), alirizacetin@selcuk.edu.tr (A.R. Cetin), ofarpag@hotmail.com (O.F. Arpag), bozturk75@hotmail.com (B. Ozturk).

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physical/mechanical properties and an increase in the elution of monomers, as well as negatively affecting the pulp tissue [2–4]. It has been found in various studies that a significant amount of organic compound residue remains unbound in the cured material [1,5,6].

Numerous studies have indicated that various components might be released from composite restorations into the oral environment [3,6,7]. Some of these substances have resulted in cytotoxic [8], mutagenic [9], genotoxic [10], and estrogenic [11] effects and pulpal, gingival, and oral mucosa reactions [12].

The final DC depends mainly on intrinsic factors such as the chemical structure of the monomer and photo initiator concentration and extrinsic factors such as polymerization conditions [13]. The energy of the light emitted from a light curing unit decreases drastically when transmitted through a resin composite, leading to a gradual decrease in the DC of the resin composite [14]. Thus far, an incremental layering technique has been the standard to sufficiently convert monomers [15]. Typically, this technique consists of placing increments of resin composite material, in a thickness of 2 mm or less, followed by exposure to light curing, and then repeating the increments until the preparation is filled [16]. One obvious advantage of this technique is limitation of the thickness of the resin to be penetrated by light. Therefore, limiting the thickness of the increments provides adequate light penetration and subsequent polymerization. Despite these benefits, there are various disadvantages associated with incremental techniques, such as the possibility of incorporation voids or contamination between composite layers, bond failures between increments, placement difficulty due to limited access in small cavities, and the increased time required to place and polymerize each layer [17].

To overcome these disadvantages, "bulk fill" composites have been introduced. These materials are claimed to promote light transmittance to enable the achievement of a reported depth of cure in excess of 4 mm [18,19]. While there is only one study in the literature investigating the clinical performance of bulk fill composites [20], investigations of marginal quality [21], cuspal deflection [19], cuspal deflection in conjunction with microleakage [22], adhesion to cavity-bottom dentin [18], and DC [23,24], post cure depth [25] have been reported. However, no studies have investigated the unreacted monomer elution from bulk fill composites.

Of the qualitative and quantitative methods of analyzing unreacted monomers and degradation products, which include gas chromatography [26], high-performance liquid chromatography (HPLC) [6], gas chromatography/mass spectrometry [27], and electrospray ionization/mass spectrometry [28], HPLC is the technique used most often.

The purpose of this study was to evaluate the elution of Bis-GMA, TEGDMA, HEMA, and Bis-EMA monomers from six bulk fill composite resins over four different time periods, using HPLC.

The null hypothesis tested were: (i) that after polymerization of a bulk fill composite resin, there would be an elution of the residual monomers in the solution, (ii) the amount of eluted residual monomers would increase with time.

2. Materials and methods

Six different composite resin materials were used in the present study: Tetric EvoCeram Bulk Fill (TF) (Ivoclar Vivadent, Amherst, NY), X-tra Fill (XF) (VOCO, Cuxhaven, Germany), SonicFill (SF) (Kerr, Orange, CA), Filtek Bulk Fill (FF) (3M ESPE Dental Products, St. Paul, MN), SDR (Dentsply, Konstanz, Germany), and EQUIA (EF) (GC America, Alsip, IL). Detailed information on the composition of the composite materials and the manufacturers are given in Table 1.

2.1. *Preparation of samples*

Six groups ($n = 5$) were formed, using each of the six composite resin materials. The samples were prepared in teflon molds, which enabled the production of standardized cylindrical specimens (4 mm thickness, 5 mm diameter). The molds were filled with the respective composite resin materials, and the samples were built up in one increment (4 mm). After the materials were inserted into the discs, a glass slide was placed on top in order to ensure smooth surfaces by extruding the excess composite resin material through the application of pressure, thereby minimizing the inhibition of the polymerization reaction by oxygen. The composite resins samples were polymerized for 20 s with a light-emitting diode unit (VALO, Ultradent, South Jordan, UT) under the standard curing mode output wavelength range 395–480 nm; output irradiance was 1000 mW/cm^2. A calibrated radiometer system (Blast LED Light Meter, First Medica, Greensboro, NC) was used to verify the irradiance at each use of the light cure unit.

One hundred and twenty total samples were produced in this manner. After fabrication, each sample was immediately immersed in 75 wt% ethanol/water solution used as extraction fluid and stored in amber-colored bottles at room temperature. Ethanol/water samples (0.5 mL) were taken for HPLC analysis at predefined time intervals: 10 min (T1), 1 h (T2), 24 h (T3), and 30 days (T4).

2.2. *Analysis*

Elutes of the specimens were analyzed by HPLC. Analytical standards, which were used for the calibration of HPLC system, were obtained from Dentsply DeTrey GmbH (Kostanz, Germany). With the exception of the reference standards, all chemicals used (ethanol and acetonitrile) were of liquid chromatographic grade. Standard solutions were prepared by dilution in 75% ethanol–water solution to achieve the required concentrations and were stored in a refrigerator (+4 °C). Intermediate (1000 μg/mL) and working standard solutions (5, 10, 25, 50 and 100 μg/mL) of each tested monomer were daily prepared with appropriate dilutions of the stock standard solution. The water was produced by an ultrapure (18.2 MΩ cm at 25 °C) purification system from Millipore (Bedford, MA). Diluted samples were passed through a 0.45 μm membrane filter prior to injection. Chromatographic analyses were performed with an Accela HPLC system that included a thermo diode array detector (DAD) and auto sampler (Thermo Fisher Scientific, San Jose, CA). Thermo Xcalibur software version 2.2 (Thermo Fisher Scientific, San Jose, CA) was used for