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Original Research Article

Elution study of unreacted TEGDMA from bulk-fill composite (SDR™ Dentsply) using HPLC

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ABSTRACT

Purpose: The study evaluates the dynamics of unreacted TEGDMA monomer elution from new generation of flowable bulk fill composite resin (SDR™ Dentsply).
Material and methods: Polymerised specimens of SDR™ composite (7 mm diameter and 4 mm thick) were placed in four solutions: 100% ethanol, 75% ethanol, distilled water and 100% methanol. The concentration of the eluted TEGDMA was measured using the HLPC method after 0.5, 1, 2 and 3 h as well as after 1, 3, 7, 14, 21 and 31 days.
Results: During the first 24 h of storage in each medium, a significant elution of TEGDMA was observed (100% ethanol – 12.5 μg/g, 75% ethanol – 8.4 μg/g, distilled water – 5.4 μg/g and 100% methanol – 7 μg/g). The elution time of the TEGDMA into 100% ethanol, 75% ethanol, distilled water and 100% methanol was 14, 7, 3 and 1 day, respectively. After 31 days, total concentrations of TEGDMA were as follows: 100% ethanol – 16 μg/g, 75% ethanol – 9.4 μg/g, distilled water – 6 μg/g and 100% methanol – 7 μg/g.
Conclusions: The TEGDMA was released from the SDR™ composite into each solution used. The TEGDMA concentration and the time of its elution depend on the type of the solvent. In an aqueous environment, the SDR™ composite exhibits a high chemical stability compared to other solutions. The direct toxicity towards to the dental pulp is established during the first hours after the placement of resin.

1. Introduction

Resin based composite (RBC) is the most widely used modern dental restorative material. It offers advantages such as excellent aesthetics and ease of handling. But it is also characterised by the risk of complications due to insufficient polymerisation of the material and the occurrence of polymerisation shrinkage. The special technique of filling cavities by using layers of composite material of up to 2 mm was developed in order to limit the above complications and to increase the durability of the fillings. It is a time-consuming procedure, especially during the filling of cavities on the occlusal or/and approximal surfaces of the posterior tooth (Class I and II restorations). One of the methods used to decrease shrinkage stress is the placement of lower density RBC on the bottom of the cavity [1–3]. In 1996, the first flowable composite was introduced into clinical practice [4]. However, mechanical properties and high polymerisation shrinkage of lower density composites did not allow for its use in thick layer [3]. Recently, a new category of flowable RBC was introduced [5,6]. In September 2009, in the USA, Dentsply launched a new type of flowable composite material called SureFil SDR Flow. In Europe, this product was introduced in February 2011 under the name of SDR™ (Smart Dentine Replacement, shrinkage decreased resin). The particularity of a new material is stated to be the option to place it in 4 mm thick bulks instead of the current incremental placement technique. Moreover, manufacturer stated that the polymerisation shrinkage of SDR™ is decreased compared to commonly used flowable and conventional RBCs. This material is indicated for use as the first, thick layer in class I and II restorations. The possibility of a rapid, single-layer application of a high amount of composite resin has changed the way large cavities are filled. In the literature, this method is described as bulk-fill technique [6–9]. The modified structure of organic resins used in SDR™ material, doubling the thickness of the polymerised composite layer and decreasing the polymerisation time compared to previous materials – raises questions concerning the impact of these parameters on the

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physiochemical properties and biocompatibility of this composite resin.

Several *in vivo* and *in vitro* studies have unequivocally demonstrated that some components of composite resins and bonding agents exhibit toxic properties [10–16]. It is believed that basic monomers: bis-GMA and UDMA, as well as TEGDMA co-monomer have high toxic potential [10,13–16]. TEGDMA, apart from the modified basic monomers UDMA and bis-EMA, is the main co-monomer of the SDR™ composite resin. TEGDMA (triethylene glycol dimethacrylate) is one of the most frequently used diluent in the composite materials. Its low molecular mass and the presence of ethylene oxide groups make this monomer reactive, mobile and relatively easy to elute from the composite material matrix [14,17–19]. The unreacted TEGDMA is a toxic substance exhibiting cytotoxic, genotoxic, mutagenic and allergenic effects [10]. It exhibits systemic and local toxicity on living organisms [10–16]. The interaction between composite materials, including TEGDMA, and exposed dental pulp is not fully known [10]. Directly capping the pulp with the use of composite resins does not lead to dentine bridge formation and may be one of the reasons for the development of inflammatory reactions in dental pulp cells, their apoptosis, as well as dental pulp inflammation and necrosis [20–22]. ED_{50} for TEGDMA, assessed in human dental pulp fibroblasts cultures, is about 0.08 mg/ml [23,24]. Therefore, similar or higher monomer concentrations, without sufficient protection of the bottom of cavity, may lead to dental pulp injures. Unreacted TEGDMA monomer may also be a substrate for micro-organisms colonising the marginal gap. It promotes the proliferation of cariogenic micro-organisms: *Lactobacillus acidofilus* and *Streptococcus sobrinus* [25].

One of the methods used to determine RBCs cytotoxicity is the unreacted monomers, released from polymerised composite resin material, concentration measurement. Based on the high-performance liquid chromatography results, it can be presumed that the cytotoxicity of the composite materials could be related to the amount of TEGDMA eluted [26–31]. Despite the fact that many authors have studied the release of residual monomers from composite materials, to date no paper assessing the release of monomers from SDR™ flow composite, polymerised in layers thicker than 4 mm, has been published.

The aim of this work was to determine the dynamics of TEGDMA monomer elution from flowable bulk fill composite resin (SDR™ Dentsply), polymerised in 4 mm layers, into four extraction medium solutions. Two null hypotheses were formulated: 1. TEGDMA monomer is released from SDR™ composite resin, and 2. The eluted monomer concentration does not depend on extraction medium type.

2. Material and methods

2.1. RBC's composition and preparation of specimens

The flowable bulk fill RBC – SDR™ (Dentsply, lot No. 384201) packaged in the form of Compula® tips, was tested. According to manufacturer' information, SDR™ flow consists of Ba-Al-F-B-Si-glass and St-Al-F-Si-glass as fillers (68% per weight, 44% per volume) and modified urethane dimethacrylate (UDMA), ethoxylated bisphenol-A-dimethacrylate (EBPADMA), triethyleneglycol dimethacrylate (TEGDMA) as resin matrix, camphoroquinone (CQ) as the photoinitiator and additives: butylated hydroxytoluene (BHT), UV stabiliser, titanium dioxide and iron oxides. Recommended polymerisation time of the 4 mm material layer is 20 s with a light intensity minimum 500 mW/cm^2.

Specimens of SDR™ composite resin (7 mm diameter and 4 mm thick) were made by placing the material into a silicone rubber mould. The surface was covered with a transparent Mylar strip. The composite was cured for 20 s using LED light curing unit: G-Light (GC). Curing was performed on one side of the sample to mimic clinical conditions. The lamp's optic fibre was in direct contact with the surface of the strip covering the material. The light intensity was 1000 mW/cm^2, and the total energy delivered to the material was 20 J/cm^2. The light intensity of the light curing unit was tested using manual radiometer. The Spring 2K Light Metre (SPR-SP3 K) manufactured by Spring Health Products Inc. Directly afterwards, the polymerised material sample were weighed using a Radwag XA82/220/X scale with an accuracy of d = 0.01/01 mg.

2.1.1. Preparation of samples for an assessment of short-term monomer elution

Sixteen discs of polymerised material were prepared and subsequently divided into 4 groups of 4 samples each. Directly after polymerisation and weighing, the sample was placed in 2 mL Eppendorf® tubes and covered with 0.5 mL of selected extraction medium: Group A – distilled water (Direct-Q 3 UV system, Millipore), Group B – 100% ethanol (gradient grade Merck), Group C – 75% ethanol (gradient grade Merck), Group D – 100% methanol (gradient grade Merck). Then it was agitated for 30 s and placed in an Eppendorf® Thermomixer Compact at 37 °C at a speed of 300 rpm. The samples were protected against light during the whole procedure. After 30 min post polymerisation, one test-tube from each of the four groups (A, B, C and D) was taken off the thermomixer. Those samples were subsequently agitated for 30 s and centrifuged (Eppendorf Centrifuge 5804R, temperature 25 °C, rpm = 14,000, t = 5 min). After centrifugation, 250 μL of obtained supernatant was taken from each sample. The collected liquid was placed in a new 2 mL Eppendorf® tube. This sample served for measuring the concentration of the TEGDMA monomer. The samples were frozen at −30 °C. A similar procedure of sample preparation was also repeated 60, 120 and 180 min after the polymerisation of the material.

2.1.2. Preparation of samples for an assessment of long-term monomer elution

Twenty discs of polymerised material were prepared and subsequently divided into 4 groups of 5 samples each. Directly after polymerisation and weighing, the sample was placed in 2 mL Eppendorf® tubes and covered with 0.5 mL of selected extraction medium: Group A – distilled water, Group B – 100% ethanol, Group C – 75% ethanol, Group D – 100% methanol. Then the tubes were agitated for 30 s and placed in the Eppendorf® Thermomixer Compact at 37 °C at a speed of 300 rpm. Sixty minutes after polymerisation, the test tubes were removed from the thermomixer and agitated for 30 s and centrifuged (Eppendorf Centrifuge 5804R, temperature 25 °C, rpm = 14,000, t = 5 min). After centrifugation, 250 μL of the obtained supernatant was taken. The collected liquid was placed in a new 2 mL Eppendorf® tube. This sample served for measuring the concentration of the TEGDMA monomer. The samples were frozen at −30 °C. The composite discs were removed from the remaining liquid, dried using of filter paper and placed in a new Eppendorf® tube. The composite discs were covered with a new portion (0.5 mL) of selected extraction medium, agitated for 30 s and subsequently placed in the thermomixer. This procedure was repeated 24 h after the polymerisation of the samples, and subsequently 3, 7, 14, 21 and 31 days after polymerisation. During the entire procedure, the samples were protected against the light.

2.2. Preparation of samples for HLPC measurements

HLPC analyses were performed using an Agilent Technologies 1200 Series system composed of a four-channel gradient pump (G1311A) with a vacuum degassing module (G1322A) and an