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Correlation of the degree of conversion with the amount of elutable substances in nano-hybrid dental composites

Jürgen Durner[a,b,*], *Julia Obermaier*[b], *Miriam Draenert*[b], *Nicoleta Ilie*[b]

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

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

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ABSTRACT

Objectives. This study's purpose was to measure and compare the degree of conversion (DC) and the amount of elutable substances from modern resin-based composites (RBCs) as function of polymerization time. One nano-hybrid RBC based on tricyclodecane-(TCD)-urethane (Venus® Diamond) and two conventionally formulated RBCs (TetricEvo Ceram®, Filtek™ Supreme XTE) were considered.

Method. DC ($n = 5$) was investigated in real time for 5 min by Fourier transform infrared spectroscopy (ATR-FTIR) in a filling depth of 2 mm at varied irradiation times (5, 10, 20, 40 s). After storing the specimens in ethanol/water for 7 d at 37 °C the eluates were analyzed by gas chromatography/mass spectrometry. Results were statistically analyzed using a multivariate analysis ($\alpha = 0.05$) an independent *t*-test ($p < 0.05$) and a Pearson correlation analysis.

Results. In all groups increasing curing time resulted in a significant increase in DC. For TetricEvo Ceram® a high significant inverse correlation was found between DC and the amount of eluted camphorquinone (CQ, Pearson correlation coefficient = −0.88), ethylene glycol dimethacrylate (EGDMA, −0.73), 4-N,N-dimethylaminobenzoic acid ethylester (DMABEE, −0.87), triethylene glycol dimethacrylate (TEGDMA, −0.68), Tinuvin P (−0.71) and bisphenol-A-polyetheylene glycol dimethacrylate (BisEMA, −0.84). Unexpectedly DC and the amount of eluted methyl acrylate (MAA) correlated directly (0.72). In the specimens of Venus® Diamond a significant inverse correlation was found between DC and the amount of eluted CQ (−0.69) and TEGDMA (−0.50), whereas in the specimen of Filtek™ Supreme XTE DC correlated with CQ (−0.96), EGDMA (−0.70), DMABEE (−0.87), TEGDMA (−0.92) and MAA (−0.92).

Significance. This study demonstrated a strong inverse correlation between DC and elutable substances in RBCs. Both evaluation methods emphasis the importance of an adequate polymerization (20, 40 s), since short curing-times (5, 10 s) resulted in lower DC and higher amount of eluted substances with toxic potential.

* *Corresponding author at*: Walther-Straub-Institute of Pharmacology and Toxicology, Faculty of Medicine, Ludwig-Maximilians-University of Munich, Nussbaumstr. 26, 80336 Munich, Germany. Tel.: +49 89 2180 73801; fax: +49 89 7095 73842.
E-mail address: juergen.durner@med.uni-muenchen.de (J. Durner).

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1. Introduction

Methacrylate based dental restoratives are widely used in practice. For dental use it is very important that methacrylate based monomers have at least two double bonds like the Bowen monomer bisphenol-A-glycidyldimethacrylate (BisGMA) or triethylene glycol dimethacrylate (TEGDMA) to form branched polymers. In contrast to (co)monomers with one double bond which can only build linear polymer chains, (co)monomers with two or more double bonds can build a three-dimensional polymer network [1]. During the polymerization process new single bonds will be built from double bond. Apart from the formation of a three-dimensional network, the degree of conversion (DC) of the (co)monomers is also important in dentistry. The DC of (co)monomers to polymers has influence on the material properties as well as on the biocompatibility of a material (the lower the DC the higher the amount of uncured monomers and additives) [2].

The Fourier transform infrared spectroscopy (FTIR) is an established method to determine remaining double bonds [3]. It must be emphasized that the remaining double bonds are not directly related to the amount of remaining (co)monomers (Fig. 1). For measurements with FTIR both cases from Fig. 1a and b are equal. In the first case (Fig. 1a) both (co)monomers are bound to the polymer chain with a single bond resulting from opening a double bond, the second double bond remains unreacted. In the second case (Fig. 1b) the (co)monomer is not linked to a polymer chain, its two double bonds are unreacted. With FTIR in both cases two unreacted double bond can be measured.

The second case is of toxicological interest because it could be shown that these (co)monomers and other substances can be eluted from polymerized dental methacrylate based materials [4]. Bleaching can degrade the three-dimensional polymer network and therefore the amount of elutable monomers and additives increases [5]. In vitro studies have shown that some of the elutable methacrylate based monomers like BisGMA, TEGDMA, 2-hydroxyethyl methacrylate (HEMA) and methyl methacrylate (MMA) as well as elutable additives like camphorquinone (CQ) can have estrogenic, mutagenic, teratogenic and genotoxic effects [6–9]. From in vivo studies it is known that during the metabolism of BisGMA, TEGDMA and HEMA epoxides like 2,3-epoxymethacrylic acid will be formed [10–12]. Furthermore elutable substances can cause allergic contact dermatitis and asthma in e.g. dentists and their personal [13].

It is discussed that the amount of elutable substances is depending on a lot of parameters like elution medium, polymerization time and the degree of cure. In earlier studies e.g. the relationship between leachability of polymerization initiator and degree of conversion of visible light-cured artificial

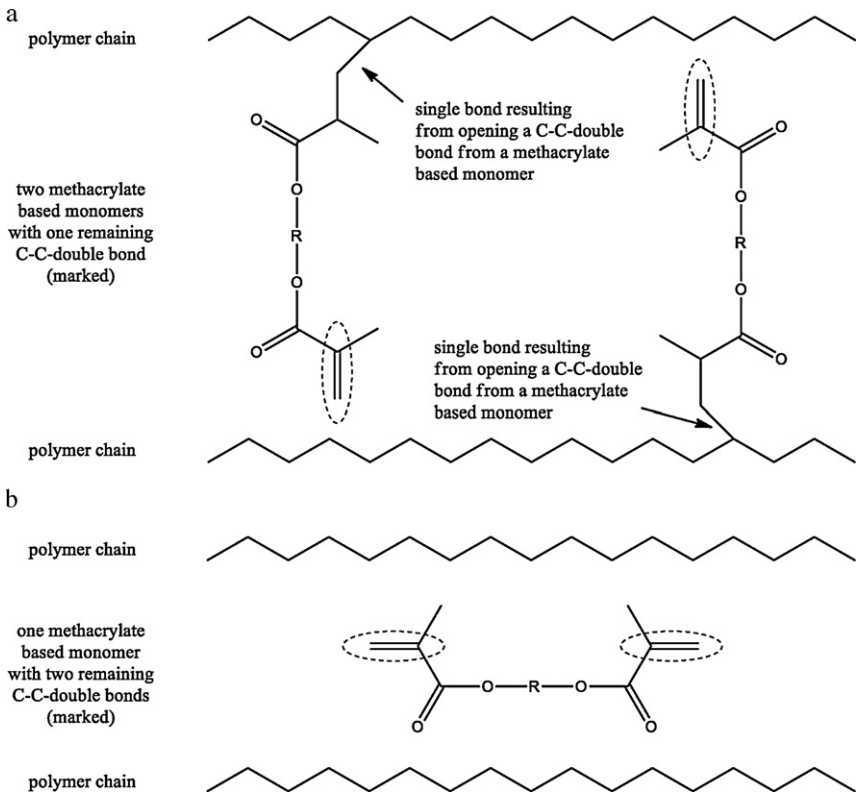

Fig. 1 – With Fourier transform infrared spectroscopy (FTIR) it is possible to detect the amount of unreacted C=C double bonds. In the case of methacrylate based monomers with two C=C double bonds it is not possible to distinguish between two cases with FTIR: (a) only one C=C double bond from the bivalent methacrylate based monomer was involved in the polymer chain formation, the other C=C double bond remains unreacted; (b) none of the C=C double bonds were involved in the polymer chain formation. Like illustrated, in both cases the amount of unreacted C=C double bonds is equal, but the consequences are not. In the second case it is possible that unreacted monomers can be eluted.