QUALITATIVE AND QUANTITATIVE ANALYSIS OF ELUTED COMPOUNDS FROM DENTAL COMPOSITES

F.-X. Reichl1,2, M. Seiss1, A. Oxynos2, M. Folwaczny1, J. Glas1, K. Kehe2, R. Hickel1
1 Department of Operative Dentistry and Periodontology, Ludwig-Maximilians-University of Munich, Goethestr. 70, 80336 Munich, Germany
2 Walther-Straub-Institute of Pharmacology and Toxicology, Ludwig-Maximilians-University of Munich, Nussbaumstr. 26, 80336 Munich, Germany, Phone: 0049/89/2180/73842; e-Mail: reichl@lmu.de

published IADR, Thessaloniki 2007

INTRODUCTION

Comonomers and monomers are used as dental restorative materials (e.g. in dental composites). Unconverted compounds can be released from dental composites and can enter the body in humans. Dental composite components can be metabolized to (toxic) intermediates in the organism.
This study was evaluated to qualify and to quantify eluted compounds from various dental composites. Following composites were tested (producer and LOT number in parentheses):
Els extra low shrinkage (Saremco; 06-2009-11), Synergy Duo Shade (Coltène; MA 015), Grandio (VOCO; 551604), Tetric Evo Ceram (Vivadent; H25200), Venus (Kulzer; 010114), Gradia (G.C.; 0502152), and Premise (Kerr; 014338).
The methods of gas chromatography/mass spectrometry (GC-MS) and liquid chromatography/mass spectrometry (LC-MS) were used.

METHODS

Polymerized composites
Polymerized composites (100 mg) were incubated in GC vials with 1 ml dest. water or 1 ml methanol, each at 37 °C for 72 hours. Aliquots were taken, and eluted compounds were analyzed with the method of gas chromatography/mass spectrometry (GC-MS) and liquid chromatography/mass spectrometry (LC-MS).

Sample analysis
Chromatographic conditions

GC-MS-Analysis
GC analyses were performed using a TraceGC Ultra-System (Thermo, Dreieich, Germany), coupled to a Thermo DSQ-mass spectrometer (Thermo, Dreieich, Germany, see figure 1).
Column: Factor Four capillary column VF-5MS (Varian, Darmstadt, Germany),
Length = 30 m, I.D. = 0.25 mm, df = 0.25 µm,
Carrier gas: Helium (flow rate 1 ml/min),
Temperature injector: 250 °C,
Temperature program: 40 °C (3 min isotherm) to 250 °C (15 min isotherm) with 12 °/min.
MS: Full-scan (m/z 50-650)

LC-MS-Analysis
LC analyses were performed using a LCQ Advantage MS-system (Thermo, Dreieich, Germany), coupled to a Finnigan Surveyor HPLC system (see figure 1).
Column: OnyxTM C18 Monolith column (50 mm length x 4.6 mm I.D.; Phenomenex, Aschaffenburg, Germany).
Mobile phase: methanol/water (0.1 % ammonium acetate).
Gradient: 10 % to 55 % methanol within 10 min, increased to 85 % methanol within 10 min and held for further 7.5 min, subsequently decreased to 10 % methanol and rinsed for further 15 min for the preparation of the next injection.
MS: Full-scan (m/z 50-650).

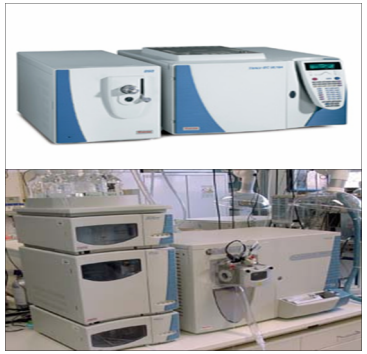

Figure 1:
Used gas chromatography/mass spectrometer (GC-MS; upper figure) and liquid chromatography/mass spectrometer (LC-MS; lower figure).

RESULTS

From all composites 18 different chemical compounds were found. Following comonomers were quantified (µg/ml; mean ± s.d.; n=3)(see table 1):

Methanol-Eluates
HEMA: The highest HEMA concentration was found in the eluate from the composite „Gradia“ (G.C.) 500 ± 66 µg/ml.
TEGDMA: The highest TEGDMA concentration was found in the eluate from the composite „Synergy Duo Shade” (Coltène) 126 ± 23 µg/ml.

Water-Eluates
HEMA: HEMA was not detectable (below limit of detection) in the eluate of any composite.
TEGDMA: The highest TEGDMA concentration was found in the eluate of the composite „Venus“ (Kulzer) 126 ± 23 µg/ml.

Additives and other compounds in the eluates (water, methanol)
Following additives were found in the range of
3 - 334 µg/ml from various composites:
DMABEE, TINP, HQME, BPE, BHT, HMBP, DCHP, TPSb, DMABEHE, DMABBEE a.o.
Isobornylmethacrylate, BisGMA, and UDMA were not found in the eluates (water, methanol) from all composites.

Table 1: Detected comonomers

	detected comonomers (µg/ml)			
composite	dest. water		methanol	
	HEMA	TEGDMA	HEMA	TEGDMA
Els extra low shrinkage	n.d.*	n.d.*	n.d.*	n.d.*
Synergy Duo Shade	n.d.*	104 ± 16	n.d.*	126 ± 23
Grandio	n.d.*	36 ± 5	n.d.*	68 ± 12
Tetric Evo Ceram	n.d.*	57 ± 12	496 ± 77	n.d.*
Venus	n.d.*	197 ± 26	n.d.*	76 ± 7
Gradia	n.d.*	123 ± 18	500 ± 66	62 ± 2
Premise	n.d.*	48 ± 7	n.d.*	51 ± 9

* n.d. = not detectable (below limit of detection)

Table 2: Abbreviations and effects of some quantified compounds

Abbreviation	Name	Effect
DMABEE	Campherchinon	Photoinitiator
TINP	4-N,N-Diethylaminobenzoacid -(2-ethylhexyl) -ester	Coinitiator
HQMP	Hydrochinonmonomethylether	Inhibitor
BPE	Benzoacidphenylester	Additive
BHT	2,6-Di-*t*-butyl-4-methylphenol	Inhibitor
HMBP	2-Hydroxy-4-methoxybenzophenon	Photostabilizer
DCHP	Dicyclohexylphthalat e	Additive
TPSb	Triphenylstiban e	Relevance ?

DISCUSSION

Different quantities of organic compounds, eluted from various composites were found. The toxicity of the eluted comonomers HEMA and TEGDMA is described in detail (Reichl *et al*. 2006, Arch. Toxicol., 80(6):370-377).
The toxicity of the eluted and detected additives (e.g. cointiators, inhibitors) is also described, but in general they have a low toxicity and for the risc assessment of dental restorative materials they are of minor relevance.

From all tested composites HEMA and TEGDMA were below limit of detection as well as in the water- and in the methanol-eluates from Els extra low shrinkage.

CONCLUSION

Following range of the eluted and detected comonomers from dental composites was found (dest. water; decreasing elution):

Venus > Gradia > Synergy Duo Shade > Tetric Evo Ceram > Premise > Grandio > Els extra low shrinkage.