Journal of Dentistry xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Journal of Dentistry

journal homepage: www.elsevier.com/locate/jdent

Curing potential of experimental resin composites with systematically varying amount of bioactive glass: Degree of conversion, light transmittance and depth of cure

Matej Par[a,*], Nika Spanovic[b], Ruza Bjelovucic[c], Hrvoje Skenderovic[d], Ozren Gamulin[e,f], Zrinka Tarle[a]

[a] Department of Endodontics and Restorative Dentistry, School of Dental Medicine, University of Zagreb, Gunduliceva 5, Zagreb, Croatia
[b] Community Health Center, Zagreb, Croatia
[c] Private Dental Practice, Sisak, Croatia
[d] Institute of Physics, Bijenicka cesta 46, Zagreb, Croatia
[e] Department of Physics and Biophysics, School of Medicine, University of Zagreb, Salata 3b, Zagreb, Croatia
[f] Center of Excellence for Advanced Materials and Sensing Devices, Research Unit New Functional Materials, Bijenicka cesta 54, Zagreb, Croatia

ARTICLE INFO

Keywords:
Bioactive glass 45S5
Degree of conversion
Light transmittance
Methacrylate polymerization
Resin composites
Bioactive composites

ABSTRACT

Objectives: The aim of this work was to investigate the curing potential of an experimental resin composite series with the systematically varying amount of bioactive glass 45S5 by evaluating the degree of conversion, light transmittance and depth of cure.

Methods: Resin composites based on a Bis-GMA/TEGDMA resin with a total filler load of 70 wt% and a variable amount of bioactive glass (0–40 wt%) were prepared. The photoinitiator system was camphorquinone and ethyl-4-(dimethylamino) benzoate. The degree of conversion and light transmittance were measured by Raman spectroscopy and UV–vis spectroscopy, respectively. The depth of cure was evaluated according to the classical ISO 4049 test.

Results: The initial introduction of bioactive glass into the experimental series diminished the light transmittance while the further increase in the bioactive glass amount up to 40 wt% caused minor variations with no clear trend. The curing potential of the experimental composites was similar to or better than that of commercial resin composites. However, unsilanized bioactive glass fillers demonstrated the tendency to diminish both the maximum attainable conversion and the curing efficiency at depth.

Conclusions: Experimental composite materials containing bioactive glass showed a clinically acceptable degree of conversion and depth of cure. The degree of conversion and depth of cure were diminished by bioactive glass fillers in a dose-dependent manner, although light transmittance was similar among all of the experimental composites containing 5–40 wt% of bioactive glass.

Clinical significance: Reduced curing potential caused by the bioactive glass has possible consequences on mechanical properties and biocompatibility.

1. Introduction

The need to reduce the incidence of secondary caries as one of the primary causes of failure of contemporary composite restorations motivates an extensive research of remineralizing composites [1–5]. Various experimental resin composites based on different ion-releasing fillers have been developed with promising results [3,5–7]. Experimental composites based on bioactive glass (BG) offer multiple benefits, originating from their potential to release remineralizing ions [8], precipitate hydroxyapatite [9], reduce bacterial penetration in the marginal gap [4], remineralize collagen fibers within the hybrid layer [10] and possibly improve the longevity of the dentin bonding [11].

The degree of conversion (DC) should be investigated in experimental composite formulations, as it is a fundamental variable which underlies their mechanical properties and potential toxicity [12,13]. The DC attainable by free-radical polymerization of composites based on bifunctional methacrylates is mainly determined by factors which affect the resin mobility throughout the polymerization: resin

* Corresponding author.
E-mail addresses: mpar@inet.hr (M. Par), spanovic.nika5@gmail.com (N. Spanovic), ruza.bjelovucic@gmail.com (R. Bjelovucic), hrvoje@ifs.hr (H. Skenderovic), ozren@mef.hr (O. Gamulin), tarle@sfzg.hr (Z. Tarle).

https://doi.org/10.1016/j.jdent.2018.06.004
Received 1 December 2017; Received in revised form 11 May 2018; Accepted 14 June 2018

composition [14], filler particle size and loading [15], and filler surface treatment [16]. The DC of light curable resin composites is always depth-dependent due to the attenuation of the curing light by light scattering at the filler/resin interface and light absorption by photoinitiator and other pigment molecules [17]. Among these phenomena, the highest attenuating effect is due to the light scattering, which is determined by filler particle size, filler load and the difference in refractive indices between the filler and resinous matrix [18]. Besides these well-known DC-determining factors, a possible inhibitory effect of unsilanized BG particles on the free radical polymerization has been suggested in preliminary studies [19,20]. Although the literature data reports the DC of experimental BG-based composites comparable to that of commercial composites [21–24], a systematic study on the relationship of the DC and BG filler load has not been published to date.

The aim of this study was to prepare a series of experimental composites with the systematically varying amount of BG (0–40 wt%) and investigate their potential to be light cured to a sufficient layer thickness in clinically acceptable time. The DC was assessed as a function of the BG amount, layer thickness and curing time. In addition, the light transmittance was evaluated and related to the thickness-dependent change of the DC. The potential to maintain a sufficient cure throughout a certain thickness was investigated by assessing the parameter "depth of cure" (DoC), which was evaluated in two ways: (I) as the layer thickness at which the DC amounts to 80% of the maximum attainable DC, and (II) as the half-length of the cylindrical composite specimen according to the classical ISO 4049 scraping test [25].

The null-hypotheses were that: (I) the DC of experimental composites is not affected by the BG amount, layer thickness or curing time; (II) the DC decline with higher layer thickness is not affected by the light transmittance; and (III) the DoC of experimental composites is at least 2 mm.

2. Materials and methods

2.1. Preparation of experimental composites

Five experimental resin composites with varying ratios of BG 45S5 (product name: G018-144, Schott, Mainz, Germany) and reinforcing fillers were prepared. The amount of BG was varied between 0–40 wt%, with reinforcing fillers up to the total filler amount of 70 wt% (Table 1). Monomers and components of the photoinitiator system were mixed in a lightproof bottle using a magnetic stirrer for 48 h. The prepared resin system was mixed with fillers in lightproof containers in a dual asymmetric centrifugal mixing system (Speed Mixer TM DAC 150 FVZ, Hauschild & Co. KG, Hamm, Germany) at 2700 rpm. The mixing was performed in five one-minute intervals separated by one-minute breaks in order to avoid overheating [26]. The composite pastes were then deaerated in vacuum for 12 h. In addition to the experimental materials, a commercial resin composite (Tetric EvoCeram, shade A2, LOT: V40834, EXP: 10/13/2020, Ivoclar Vivadent, Schaan, Liechtenstein, filler load: 76 wt%/54 vol%, abbreviation: TEC) was used as a reference.

2.2. Degree of conversion

The DC was assessed using the previously developed setup for repeated measurements at different points of cylindrical specimens (d = 3 mm, h = 5 mm), simulating different layer thicknesses [26]. The composite paste was applied into a custom-made stainless steel split-mold, the mold apertures were covered with a polyethylene terephthalate (PET) strip and curing was performed for 20 s or 40 s through the upper aperture with a light-emitting diode (LED) curing unit (Bluephase G2, Ivoclar-Vivadent, Schaan, Liechtenstein; wavelength range 380–515 nm, nominal irradiance of 1200 mW/cm^2). The curing unit tip was positioned perpendicularly to the mold aperture, immediately adjacent to the PET strip. The environmental temperature during curing was 21 ± 1 °C. Immediately after curing, Raman spectra were recorded from the upper specimen surface (denoted as the thickness of 0 mm). After the measurement, the specimens were transferred to an incubator (Cultura, Ivoclar-Vivadent, Schaan, Liechtenstein) for dark storage at 37 ± 1 °C. After 24 h post-cure, Raman spectra were collected from five precisely determined points on the cylindrical specimen, representing layer thicknesses of 0, 1, 2, 3 and 4 mm. For each experimental group, five specimens were prepared (n = 5).

FT-Raman spectroscopy measurements were performed using a Spectrum GX spectrometer (PerkinElmer, Waltham, USA) with the excitation NdYaG laser of 1064 nm wavelength, the laser power of 800 mW and resolution of 4 cm^{-1}. The laser was targeted at the most prominent part of the cylindrical specimen surface which was positioned in the collecting lens focus. The diameter of the excited area was 0.5 mm and 50 scans were recorded for each spectrum (12 s per scan). Spectra of the uncured composites (n = 5) were recorded in the same manner. Raman spectra were processed with the Kinetics add-on for Matlab (Mathworks, Natick, Massachusetts, USA).

The DC was calculated by comparing the relative change in height of the spectral bands at 1640 cm^{-1} (aliphatic C═C) and 1610 cm^{-1} (internal standard, aromatic C═C) before and after the curing, according to the equation [27]:

$$DC = 1 - \frac{(1640\,cm^{-1} / 1610\,cm^{-1})_{peak\ height\ after\ curing}}{(1640\,cm^{-1} / 1610\,cm^{-1})_{peak\ height\ before\ curing}}$$

Table 1
Composition of the experimental resin composites.

Material		Filler composition (wt%)		Total filler ratio (wt%)	Resin		Filler load (vol%)
		Bioactive glass	Reinforcing fillers (Ba:Si = 2:1)		wt%	Composition	
non-bioactive control	**BG-0**	0	70	70	30	60% Bis-GMA	48
BG-containing composites	**BG-5**	5	65	70	30	40% TEGDMA	48
	BG-10	10	60	70	30	photoinitiator system:	48
	BG-20	20	50	70	30	0.2% CQ	51
	BG-40	40	30	70	30	0.8% 4E	52

Bioactive glass: SiO_2 45%, Na_2O 25%, CaO 25%, P_2O_5 5%, particle size (d50/d99 [μm]): 4.0/13.0, silanization: none, product name/manufacturer: G018-144/Schott, Germany.
Barium-fillers (Ba): SiO_2 55.0%, BaO 25.0%, B_2O_3 10.0%, Al_2O_3 10.0%, particle size (d50/d99 [μm]): 1.0/4.0, silanization 3.2 wt%, product name/manufacturer: GM27884/Schott, Germany.
Silica-fillers (Si): SiO_2 ≥ 99.8%, primary particle size: 12 nm, silanization 4–6 wt%, product name/manufacturer: Aerosil DT/Evonik Degussa, Germany.
Bis-GMA: Bisphenol A glycidyl methacrylate, Esstech, PA, USA; TEGDMA: tri-ethylene glycol dimethacrylate, Esstech; CQ: camphorquinone, Aldrich, WI, USA; 4E: ethyl-4- (dimethylamino) benzoate, Aldrich.