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Network structures of Bis-GMA/TEGDMA resins differ in DC, shrinkage-strain, hardness and optical properties as a function of reducing agent

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ABSTRACT

Objectives. To evaluate the influence of different tertiary amines on degree of conversion (DC), shrinkage-strain, shrinkage-strain rate, Knoop microhardness, and color and transmittance stabilities of experimental resins containing BisGMA/TEGDMA (3:1 wt), 0.25 wt% camphorquinone, 1 wt% amine (DMAEMA, CEMA, DMPT, DEPT or DABE). Different light-curing protocols were also evaluated.

Methods. DC was evaluated with FTIR-ATR and shrinkage-strain with the bonded-disk method. Shrinkage-strain-rate data were obtained from numerical differentiation of shrinkage-strain data with respect to time. Color stability and transmittance were evaluated after different periods of artificial aging, according to ISO 7491:2000. Results were evaluated with ANOVA, Tukey, and Dunnett's T3 tests ($\alpha = 0.05$).

Results. Studied properties were influenced by amines. DC and shrinkage-strain were maximum at the sequence: CQ < DEPT < DMPT ≤ CEMA ≈ DABE < DMAEMA. Both DC and shrinkage were also influenced by the curing protocol, with positive correlations between DC and shrinkage-strain and DC and shrinkage-strain rate. Materials generally decreased in L^* and increased in b^*. The strong exception was the resin containing DMAEMA that did not show dark and yellow shifts. Color varied in the sequence: DMAEMA < DEPT < DMPT < CEMA < DABE. Transmittance varied in the sequence: DEPT ≈ DABE < DABE ≈ DMPT ≈ CEMA < DMPT ≈ CEMA ≈ DMAEMA, being more evident at the wavelength of 400 nm. No correlations between DC and optical properties were observed.

Significance. The resin containing DMAEMA showed higher DC, shrinkage-strain, shrinkage-strain rate, and microhardness, in addition to better optical properties.

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1. Introduction

Significant improvements in the formulation of resin-composites have been achieved since light initiation was introduced. Since their introduction as a restorative material in the early 1960s, the development of these materials has resulted in better physical, mechanical, and biological properties. For this reason, resin-composites have been widely employed in both anterior and posterior teeth as well as in direct and indirect restorations.

It is known that the properties of resin-based materials are directly related to the filler content [1–5] as well as the degree of conversion [6]. An adequate degree of conversion (DC) is important to guarantee not only strength and esthetics, but also biocompatibility [7]. When insufficient conversion occurs, higher concentrations of unreacted monomers are released to the oral environment. Although all components of resin-composites may leach out in aqueous solutions [8,9], the release of unreacted monomers is a concern because of their cytotoxic potential [7]. Additionally, tertiary amines from the initiator system may induce pulpal cells apoptosis [10].

Several factors may influence the DC, including features of the light-curing unit (such as power density, light spectrum, and exposure time), and the composition of the material [11–15]. The shade of the resin-composite and the type and concentration of light-initiator or reducing agent used in the formulation may also influence the final quality of the polymerization reaction [16–18]. Another important factor that may influence the properties of resin-based materials is the viscosity [19–21]. When different resin comonomers are evaluated, the viscosity of the system plays an important role in the polymerization process. Mixtures containing between 50 and 75 wt% of BisGMA display the higher rates of polymerization [22]. On the other hand, if the increase in the DC improves mechanical properties of resin-composites [18,23], it has the disadvantage of increasing the contraction stresses, as a direct relationship between the DC and the contraction stresses has been observed [12,24,25]. These contraction stresses can damage the tooth–restoration interface or even cause cuspal deflection [26,27].

The most common light-initiator used in dental resins is camphorquinone (CQ), while the reducing agents, the tertiary amines, may vary. The type of co-initiator present in the photo-initiator system plays an important role in determining the physical-chemical properties of resin-composites. Different tertiary amines, such as DMAEMA (2-dimethylaminoethyl methacrylate), CEMA (N,N-cyanoethylmethylaniline), DMPT (NN-dimethyl p-toluidine), DEPT (N,N-diethanol p-toluidine), and DABE (N,N-dimethyl-p-aminobenzoic acid ethylester), can be associated with the camphorquinone in the light initiation of resin-based materials [11,15,21,23,28–32].

When a resin-composite restoration is prepared, it is expected that there will be not only to have good mechanical properties, but also stable optical properties. The color of a resin-composite may be affected not only by staining from exogenous sources, such as coffee, tea, or red wine, but also by chemical differences of resin components, such as polymeric structure, photo-initiator system, and filler particles size and distribution [16,29,33]. Although several studies on color stability of resin-composites have been reported, the influence of different co-initiators on DC, shrinkage, hardness, and color in experimental materials, without considering the influence of fillers, has not yet been confirmed. Thus, it is the aim of the present study to evaluate the DC, shrinkage-strain, shrinkage-strain rate, Knoop microhardness, and color and transmittance stabilities of experimental resins containing different amine co-initiators. It is hypothesized that different tertiary amines in experimental resins would influence the degree of conversion, the shrinkage-strain and shrinkage-strain rate, and the Knoop microhardness. It is also hypothesized that both color and transmittance stabilities would be influenced by different tertiary amines when cured resins are exposed to artificial daylight aging.

2. Materials and methods

2.1. Preparation of resins

Experimental fluid resin-monomers mixtures containing BisGMA and TEGDMA comonomers at the ratio of 3:1 by mass, respectively, with 0.25 mass% camphorquinone (CQ) and 1.0 mass% amine were formulated. Different tertiary amines were added, each of them corresponding to a specific experimental resin (Table 1 and Figs. 1 and 2). No amine was added to the control formulation. All materials were used without further purification.

2.2. Degree of conversion

The DC was analyzed by Fourier transform infrared spectroscopy (Avatar 360 FT-IR ESP, Nicolet Analytical Instruments, UK), using an attenuated total reflectance unit (MIRacle ATR, Pike Technologies, Madison, USA) at the resolution of 4 cm^{-1} and 32 scans in the range of 4000–800 cm^{-1}. Three curing protocols were evaluated: 20 s, 40 s, and 40 s plus additional 7 min light-curing. Disk-shaped samples (10 mm × 1 mm), prepared with a Teflon mold between two 1-mm-thick glass slides, were cured with a QTH light-curing unit (Optilux 501, SDS Kerr, Danbury, USA) operating on standard mode and emitting 760 mW/cm^2 irradiance, as measured by the incorporated radiometer, which was calibrated against a flat-response power-meter. The additional curing was performed in a curing chamber (Visio Beta Vario, 3M-Espe, USA). The ratios of the infrared spectra of aliphatic (1638 cm^{-1}) to aromatic (1608 cm^{-1}) C=C double bonds absorption peaks in cured and uncured resins were used to calculate monomers conversion. Three replicates for each experimental resin were made. DC values were obtained after 0 h, 1 h, 24 h, and 168 h (7 days) storage in dark canisters at room temperature ($23 \pm 1\,^{\circ}\text{C}$).

2.3. Shrinkage-strain and shrinkage-strain rate

The shrinkage-strain was measured using the bonded-disk method [34,35] at $23 \pm 1\,^{\circ}\text{C}$ and $37 \pm 1\,^{\circ}\text{C}$. Two curing protocols were evaluated: 20 s and 40 s. Three replicates for each experimental resin were made. Shrinkage-strain rates were obtained by numerical differentiation of shrinkage-strain data