



AUREALICE ROSA MARIA MARTINS

**“INFLUENCE OF DILUENT ISOBORNYL METHACRYLATE
MONOMER (IBOMA) ON PHYSICO-MECHANICAL PROPERTIES
OF EXPERIMENTAL RESIN”**

**“INFLUÊNCIA DO MONÔMERO DILUENTE ISOBORNIL
METACRILATO (IBOMA) EM PROPRIEDADES FÍSICO-
MECÂNICAS DE RESINAS EXPERIMENTAIS”**

PIRACICABA

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UNIVERSIDADE ESTADUAL DE CAMPINAS

FACULDADE DE ODONTOLOGIA DE PIRACICABA

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MONOMER (IBOMA) ON PHYSICO-MECHANICAL PROPERTIES
OF EXPERIMENTAL RESIN”**

Orientador: Prof. Dr. Mário Alexandre Coelho Sinhoreti

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METACRILATO (IBOMA) EM PROPRIEDADES FÍSICO-
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Dedico este trabalho

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*"Para realizar grandes conquistas,
devemos não apenas agir, mas também sonhar;
não apenas planejar, mas também acreditar"*

Anatole France.

Resumo

O objetivo neste estudo foi avaliar a utilização do isobornil metacrilato (IBOMA) como monômero diluente associado ou não ao TEGDMA, em resinas experimentais à base de Bis-GMA. No Capítulo 1, o grau de conversão (GC), a resistência à flexão (RF), o módulo de elasticidade (ME), a resistência máxima à tração (RMT), a profundidade de polimerização (PP) e a contração volumétrica (CV) foram avaliados. No Capítulo 2, a dureza Knoop (KHN), a densidade de ligações cruzadas (DLC) e a sorção (SOR) foram avaliadas. Nove formulações de matriz resinosa foram confeccionadas de acordo com o monômero diluente IBOMA, TEGDMA ou IBOMA / TEGDMA) nas proporções: Bis-GMA/IBOMA 60/40, 50/50, 40/60% em peso, Bis-GMA/TEGDMA 60/40, 50/50, 40/60% e quando associados Bis-GMA/IBOMA/TEGDMA, 60/20/20, 50/25/25 e 40/30/30%). Para o GC, os espécimes em formato de barras (7 x 2 x 1 mm / n=10) foram avaliados em espectroscopia de infravermelho transformada de Fourier (FTIR). Em seguida, os testes de RF e MF foram conduzidos numa máquina de ensaios universal, na mesma amostra utilizada para a avaliação do GC. A PP foi avaliada em amostras (n=5) confeccionadas, seguindo as instruções da ISO 4049. A densidade dos espécimes antes (pu) e após (pc) a fotoativação foram utilizadas para avaliar a CV, utilizando o princípio de Arquimedes. DLC foi avaliada através do teste de amolecimento; testes de KHN foram realizados antes e depois da imersão em etanol absoluto, durante 24 horas. O teste de SOR (n=5) foi realizado de acordo com as normas da ISO 4049. Os dados foram submetidos à análise de variância de dois fatores e teste de Tukey ($\alpha=0,05$). As resinas Bis-GMA/TEGDMA apresentaram maiores valores de GC do que as resinas que continham IBOMA, exceto para a mistura com 60% em peso de Bis-GMA ($p\leq 0,001$). As resinas com IBOMA apresentaram menor RF, MF e RMT do que outros grupos ($p\leq 0,001$). A PP para as misturas de IBOMA, foi menor do que para os outros grupos ($p\leq 0,001$), exceto para as resinas que continham elevada concentração de monômero diluente (Bis-GMA/TEGDMA, 40/60% em peso). As resinas Bis-GMA/ TEGDMA 60/40% e Bis-GMA/IBOMA/TEGDMA 60/20/20% apresentaram os valores mais elevados de dureza Knoop e as resinas Bis-GMA/IBOMA 50/50% e Bis-GMA/IBOMA 40 / 60% apresentaram os menores valores DLC. As resinas que continham TEGDMA apresentaram maior SOR. Pode-se concluir que o IBOMA diminuiu a contração volumétrica. No entanto, os copolímeros que contêm IBOMA sozinho

apresentaram menores propriedades físico-mecânicas do que os sistemas Bis-GMA/TEGDMA e Bis-GMA/TEGDMA/IBOMA.

Palavra-chave: monômero, IBOMA, Bis-GMA, TEGDMA, contração volumétrica

Abstract

The purpose in this study was to evaluate the use of isobornyl methacrylate IBOMA as diluent monomer in the Bis-GMA/TEGDMA blends. In the Chapter 1 the degree of conversion (DC), flexural strength (FS), elastic modulus (EM), maximum tensile strength (MTS), depth of cure (DCU) and volumetric shrinkage (VS) were evaluated; In the Chapter 2 the Knoop hardness (KHN), crosslink density (CLD) and sorption (WS) were evaluated. Nine resin matrix formulations were made according to diluent monomer IBOMA, TEGDMA or IBOMA/TEGDMA in the following proportions: Bis-GMA/IBOMA 60/40, 50/50, 40/60 wt%, Bis-GMA/TEGDMA 60/40, 50/50, 40/60 wt% and when associated Bis-GMA/IBOMA/TEGDMA, 60/20/20, 50/25/25 e 40/30/30 wt%. DC was evaluated in bars (7 x 2 x 1 mm/ n=10) by Fourier transformed infrared spectroscopy (FTIR). FS and FM tests were conducted in a universal test machine in the same sample used to DC evaluation. DCU was evaluated in samples (n=5) made following the ISO 4049:2009 directions. The densities of samples before (pu) and after (pc) photoactivation were used to evaluate the VS using the Archimedes principle. CLD was evaluated through softening test; KHN tests were performed before and after immersion in absolute ethanol for 24 h. WS test (n = 5) was performed following the ISO 4049:2009 directions. Data were submitted to two-way ANOVA and Tukey's test ($\alpha=0.05$). The resins Bis-GMA/TEGDMA showed higher DC than resins containing IBOMA, except for the blend with 60 wt% Bis-GMA ($p\leq 0.001$). The blends with IBOMA showed lower FS, EM and MTS than other groups ($p\leq 0.001$). DCU for IBOMA blends was lower than other groups ($p\leq 0.001$), except for the highest concentration of diluent monomer (Bis-GMA/TEGDMA, 40/60 wt%). The resins Bis-GMA/TEGDMA 60/40% and Bis-GMA/IBOMA/TEGDMA 60/20/20% showed the highest KHN values of and resins Bis-GMA/IBOMA 50/50% and Bis-GMA/IBOMA 40/60% showed the lowest CLD values. The resins containing TEGDMA showed higher WS. It can be concluded that IBOMA decrease the volumetric shrinkage. However, copolymers containing IBOMA alone showed lower physical-mechanical properties than Bis-GMA/TEGDMA and Bis-GMA/IBOMA/TEGDMA systems.

Keyword: monomer, Bis-GMA, TEGDMA, IBOMA, volumetric shrinkage

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INTRODUÇÃO

As resinas compostas odontológicas são materiais restauradores de grande importância na Odontologia e foram introduzidos por Bowen em 1962, sendo que, ainda hoje, geram preocupações devido aos processos deletérios gerados pela tensão causada durante a reação de polimerização (GONÇALVES *et al.*, 2010). A maioria dos materiais resinosos disponíveis no mercado depende de composições à base de monômeros metacrilatos, devido à elevada resistência mecânica, baixa volatilidade e contração de polimerização relativamente baixa (PEUTZFELDT, 1997), os quais irão reagir entre si, formando cadeias poliméricas de alto peso molecular. A tensão causada na interface de união durante a reação de polimerização pode favorecer o insucesso das restaurações induzindo a formação de fenda, sensibilidade pós-operatória e microinfiltração /pigmentação marginal (FERRACANE *et al.*, 2005; GONÇALVES *et al.*, 2008; PFEIFER *et al.*, 2008; STANSBURY *et al.*, 2005; MANHART *et al.*, 2004; VERSLUIS *et al.*, 2004). Sabe-se que alto grau de conversão pode estar associado ao elevado módulo de elasticidade e à contração volumétrica do compósito, e ambos estão relacionados com a tensão de polimerização (GONÇALVES *et al.*, 2008). O grau de conversão elevado é, no entanto, altamente desejável para proporcionar a resistência mecânica necessária para o material suportar cargas mastigatórias e degradações químicas no ambiente bucal (VANDEWALLE *et al.*, 2004; BENETTI *et al.*, 2009). Portanto, o desenvolvimento de materiais com combinação de alto grau de conversão, baixa contração e baixa tensão de polimerização é desejável, e ainda permanece um desafio.

Com o intuito de solucionar ou pelo menos minimizar esses problemas, novas técnicas de aplicação e novos materiais vêm sendo desenvolvidos. Em relação ao desenvolvimento de materiais, algumas novas formulações para a matriz das resinas compostas vêm sendo estudadas, visando sintetizar monômeros com melhores propriedades físicas e mecânicas (FRÓES-SALGADO *et al.*, 2011; HE *et al.*, 2011; LABELLA *et al.*, 1998;). O monômero-base mais utilizado atualmente nos compósitos odontológicos é o bisfenol glicidil metacrilato (Bis-GMA), por ser pouco volátil, ter baixa difusividade entre os tecidos e baixa contração volumétrica (MAHMOODIAN *et al.*, 2008). No entanto, é altamente viscoso à temperatura ambiente (KHATRIA *et al.*, 2003) e geralmente está associado a monômeros diluentes para permitir não só a incorporação de carga inorgânica,

mas também para melhorar a mobilidade dos monômeros que compõem a matriz durante a reação de polimerização, aumentando o grau de conversão. A conversão mais elevada, aliada à concentração aumentada de grupos reativos, leva ao aumento da contração volumétrica; no entanto, contribui para redes mais reticuladas e densas, formando polímeros menos propensos à degradação (PEUTZFELDT *et al.*, 1997).

Assim, dimetacrilatos menos viscosos, tais como o trietilenoglicol dimetacrilato (TEGDMA), foram adicionados, de 20 a 50% em peso para melhorar o manuseio das formulações das resinas dentárias, bem como para alcançar graus elevados de conversão (PEUTZFELDT *et al.*, 1997). O TEGDMA é um dos monômeros diluentes mais adicionados à matriz com monômero de alto peso molecular, pois tem baixa viscosidade e favorece o aumento do grau de conversão (STANSBURY *et al.*, 2001; ODIAN, 1991); no entanto, apresenta alta contração volumétrica (GONÇALVES *et al.*, 2008). Ainda, a adição do diluente TEGDMA em maior proporção, aumenta a contração de polimerização e sorção de água pela matriz (KHATRIA *et al.*, 2003). BURKE *et al.* (1999) verificaram que resinas compostas à base de Bis-GMA/TEGDMA são as que mais apresentam falha prematura de união com a estrutura dentária. Muitas alternativas de monômeros diluentes com intenção de substituição parcial ou total do TEGDMA foram exploradas como meio para reduzir esses problemas (NIE *et al.*, 2001; LOVELL & BOWMAN, 2001). Monometacrilatos e acrilatos, tais como metacrilato de hidroxipropil e isobornil acrilato, têm sido estudados e se mostrado capazes de reduzir a contração volumétrica e aumentar as conversões monoméricas (LABELLA *et al.*, 1998; DAVY *et al.*, 1998).

O Isobornil acrilato (IBOA) e o isobornil metacrilato (IBOMA) são dois monômeros que apresentam baixas viscosidade e contração de polimerização. Estudos mostram a utilização de IBOA e IBOMA como diluentes em resinas para revestimento do tipo verniz (CUI *et al.*, 2007) ou como monômeros diluentes de matriz de resinas compostas, por causa da baixa viscosidade, baixo potencial de contração de polimerização e de elevada hidrofobia (HE *et al.*, 2011). Ainda, têm sido recentemente utilizados na sintetização de nanogéis para a matriz de resinas compostas, com o objetivo de reduzir a contração e consequente tensão de polimerização. (MORAES *et al.*, 2011; LIU *et al.*, 2012). Assim, a incorporação do IBOMA, como monômero diluente, poderia diminuir a contração de matrizes de resinas compostas. Baseado nisso, neste estudo o objetivo foi

avaliar o efeito do IBOMA como monômero diluente do Bis-GMA, sozinho ou em associação com TEGDMA, em diferentes proporções, sobre algumas propriedades físico-mecânicas dessas matrizes resinosas resultantes. Para tanto, essa dissertação foi confeccionada em formato alternativo, em dois capítulos, tendo como objetivo avaliar propriedades físico-mecânicas de várias formulações dessas resinas experimentais: 1) No Capítulo 1 foram avaliados grau de conversão, resistência à flexão, módulo de elasticidade, resistência máxima à tração, profundidade de polimerização e contração volumétrica; e 2) No Capítulo 2: dureza, densidade de ligações cruzadas e sorção dessas mesmas formulações.

Capítulo 1

Isobornyl methacrylate as diluent co-monomer in model dental resins: evaluation of physical-mechanical properties

Abstract

Objectives: The aim of this study was to evaluate the effect of IBOMA as a diluent co-monomer on the degree of conversion (DC), flexural strength (FS), elastic modulus (EM), ultimate tensile strength (UTS), depth of cure (DCU) and volumetric shrinkage (VS) of the experimental resins in function of the copolymer content and concentrations.

Materials and methods: A blend of Bis-GMA as base monomer was formulated with different concentrations of diluent, such as TEGDMA(60/40, 50/50 e 40/60wt%), IBOMA (60/40, 50/50 and 40/60wt%), or TEGDMA/IBOMA (60/20/20, 50/25/25 and 40/30/30 wt%). The Camphoroquinone 0.5wt%/2-(N,N-Dimethylamino)ethyl methacrylate 1wt% (CQ/DMAEMA) was used as the photoinitiator system and inhibitor butylated hydroxytoluene 0.1 wt% (BHT). The DC was evaluated by FTIR. The FS and EM were determined by three-point bending test. For the UTS, hourglass samples were tested in tension. Immediate and 24h VS were calculated using the density parameters before and after curing. Data were submitted to ANOVA and Tukey's test ($\alpha = 0.05$).

Results: Bis-GMA/IBOMA showed lower physical-mechanical properties (UTS, DCU, FS, EM, DC) but also, lower immediate and 24h VS. The results of TEGDMA as the diluent monomer in Bis-GMA resins, alone or in association with IBOMA, showed better curing performance and increased physical-mechanical properties when compared with IBOMA blends.

Conclusion: The experimental resin containing only IBOMA as diluent monomer exhibited the lower volumetric shrinkage; however, presented also lower physical-mechanical properties when compared to the resins containing only TEGDMA or the association of both.

Keywords: methacrylate monomers, dental resin, curing performance.

Introduction

Resin composites have been widely used in direct esthetic restorative procedures. However, polymerization shrinkage still harms the tooth/restoration interfacial quality, creating marginal adaptation problems [1]. The shrinkage under confinement can develop stresses on the bond interface [2], potentially leading to gap formation, marginal pigmentation, and secondary caries. The presence of gaps on the tooth/resin interface is considered the first sign of restoration failure [3].

Most of the commercial dental resins present dimethacrylate compositions, in which the organic matrix consists of a base resin and a diluent co-monomer. Usually, the bisphenol glycidyl methacrylate (Bis-GMA) is the base monomer, therefore, due to the high viscosity under ambient polymerization conditions [4], diluent monomers are added to overcome this drawbacks. In an attempt to improve the handling of dental resins and achieve a higher degree of conversion, the Bis-GMA is most commonly associated with the co-monomer triethylene glycol dimethacrylate (TEGDMA) [5,6].

Adversely, the incorporation of TEGDMA increases the volumetric shrinkage, since it contains higher carbon double bonds per volume and allows higher degree of conversion [7]. Also, Bis-GMA/TEGDMA resins generate high shrinkage stress, which can disrupt the bond interface and decrease the lifetime of the adhesive restoration [8,9]. In this sense, some alternative diluent co-monomers have been studied in order to replace the TEGDMA in dental resins and exhibit lower volumetric shrinkage stress and its determinants [7,10]

The monomethacrylate Isobornyl methacrylate (IBOMA) has been studied as diluent co-monomer used in coatings [11] and for polymer purposes [12]. Also, IBOMA is a traditional monomethacrylate that has low viscosity and low shrinkage potential [13,14]. Considering degradation resistance, this co-monomer also presents lower water sorption, which can increase the polymer durability, especially in the oral environment[13]. However, it would be interesting more studies on polymerization and mechanical behavior of resins containing IBOMA as diluent co-monomer, either regarding its concentration variance, or its performance while in association with TEGDMA.

Thus, the aim of this study was to evaluate the influence of the diluent co-monomer content and concentration on the physical-mechanical properties of experimental dental resins. The tested hypothesis was: IBOMA alone or in association with TEGDMA in Bis-

GMA based resins would exhibit lower volumetric shrinkage and similar physical-mechanical properties compared to TEGDMA.

Materials and methods

Resin preparation

Nine experimental resin formulations were tested in this study. The resin matrix consisted on bisphenol glycidyl methacrylate (Bis-GMA - Sigma-Aldrich Inc, St Louis, MO, USA) as base monomer and two diluent monomers of low molecular weight: triethyleneglycol dimethacrylate (TEGDMA - Sigma-Aldrich Inc, St Louis, MO, USA), MW 286,3; and isobornyl methacrylate (IBOMA - Sigma-Aldrich Inc, St Louis, MO, USA), MW 222,32. The diluent monomers were mixed with the base monomer in 9 different proportions, as described in Table 1. The photo-initiator system was composed of camphorquinone (CQ – 0.5 wt%, Sigma-Aldrich Inc, St Louis, MO, USA) and 2-(dimethylamino)ethyl methacrylate (DMAEMA – 1 wt%, Sigma-Aldrich Inc, St Louis, MO, USA). Also, the inhibitor butylated hydroxytoluene (BHT, Sigma-Aldrich Inc, St Louis, MO, USA) was added to the organic matrix in a concentration of 0.1 wt% to avoid spontaneous polymerization of the blends. The molecule structures of the monomers are presented in Figure 1.

Table 1. Composition of the experimental resin blends used.

Resin	Bis-GMA%	TEGDMA%	IBOMA%
Resin 1 - 60/40/0	60	40	-
Resin 2 - 50/50/0	50	50	-
Resin 3 - 40/60/0	40	60	-
Resin 4 - 60/0/40	60	-	40
Resin 5 - 50/0/50	50	-	50
Resin 6 - 40/0/60	40	-	60
Resin 7 - 60/20/20	60	20	20
Resin 8 - 50/25/25	50	25	25
Resin 9 - 40/30/30	40	30	30

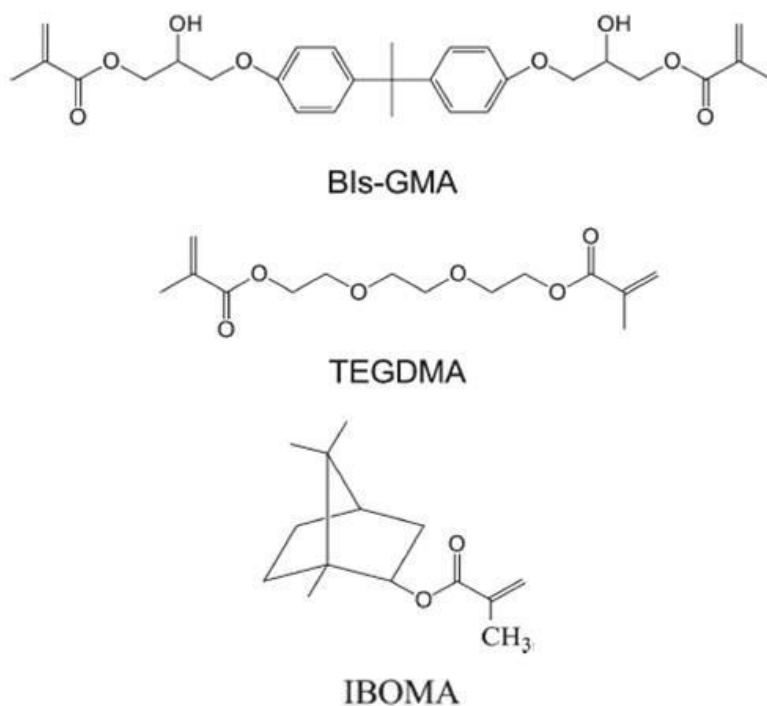


Figure 1. Bis-GMA, TEGDMA and IBOMA molecules.

Degree of conversion

For DC analysis bar-shaped samples (7mm length x 2mm width x 1mm thickness) (n=10) were made and photo-activated by a LED curing unit (Bluephase G2, 1200 mW/cm²), during 60s. The total energy dose was standardized at 72J. After polymerization, the samples were removed from the molds and dry stored in light-proof containers at 37°C, during 24 hours. DC was undertaken after this period on the top surface of each specimen using a Fourier transformed infrared spectroscopy at attenuated total reflectance (FTIR/ATR - Spectrum 100, PerkinElmer, Shelton, CA, USA)

The absorption spectra of non-polymerized and polymerized experimental resins were obtained from the region between 4000 and 650 cm⁻¹ with 32 scans at 4 cm⁻¹. For the tested resins containing aromatic vinyl bonds of bisphenol and aliphatic bonds of the methacrylate functional group, the aliphatic carbon-to-carbon double-bond absorbance peak

intensity (located at 1638 cm⁻¹) and that of the aromatic component (located at 1608 cm⁻¹; reference peak) were obtained. The DC (%) was calculated using the following equation:

$$DC (\%) = 100 \times [1 - (R_{\text{polymerized}}/R_{\text{nonpolymerized}})],$$
 where R represents the ratio between the absorbance peak at 1638 cm⁻¹ and 1608 cm⁻¹.

Flexural strength and elastic modulus

Immediately after the DC analysis, the bar-shaped samples (n = 10) were used for the three-point bending flexural test. The test was performed according to ISO 4049:2009 [15], except for the specimen's dimensions (7 mm length x 2 mm width x 1 mm thickness). The three-point bending test was performed in a universal testing machine (Instron, Canton, USA – span between supports = 5 mm) at a cross-head speed of 0.5 mm/min. The maximum load recorded for the samples fracture was recorded in Newton (N) and the FS in MegaPascals (MPa) using the following equation:

$$FS = 3FL/(2BH^2)$$

Where: F - the maximum load (N) exerted on the samples; L - the distance (mm) between the supports; B - the width (mm) of the samples; H - the height (mm) of the samples. The EM was measured as the slope of the strain x strain curve on the linear portion, using the following equation of the software Bluehill 2 (Instron Corporation, 825 University Avenue Norwood, MA, EUA).

$$E = L_1 D^3 10^{-3} / 4BH^3 D$$

where: L₁ - load (N); D - distance (mm) between the supports; B - width (mm); H - height (mm); D - displacement (mm).

Ultimate tensile strength

An hourglass shaped silicon mold (10 mm long x 0.9 mm thick, with a median cross-section of 0.9 x 0.9 mm) was employed for the construction of the samples. The experimental resins were inserted into the mold, a polyester strip was placed on the top surface and then photoactivated by LED curing unit (Bluephase G2, 1200mW/cm²), for 60s. After the photoactivation procedure, the hourglass samples were dry stored for 24h at

37°C. Then, the samples were attached to the tensile jig with cyanoacrylate adhesive (Zapit, Dental Ventures of America, Corona, CA, USA) and tested in tension in a universal testing machine (EZ-Test, Shimadzu, Kyoto, Japan), at a cross-head speed of 0.5mm/min. The UTS, expressed in MPa, was calculated as:

$$UTS = F/A$$

Where: F - tensile force (N), A - cross-sectional area of the specimen (mm²).

Depth of cure

The DCU test was performed according to ISO 4049:2009 [15] was performed. Cylindrical samples (n=5) were prepared (20 mm height x 3 mm diameter). The experimental resins were inserted into the mold. A polyester strip and an 1-mm glass slide spacer were placed on the top of the mold and then the samples were photoactivated for 60s using a LED curing unit (Bluephase G2, 1200mW/cm²). After light-curing, the cylindrical samples were removed from the mold and the soft material underneath was scrapped away using a plastic spatula. The sample remaining was measured by a digital caliper (Mitutoyo Corporation, Tokyo, Japan) with ± 0.01 mm accuracy. This value was divided by two to obtain DCU (ISO 4049:2009)

Volumetric shrinkage

The volumetric shrinkage (VS) was determined by measuring the resin density before (ρ_u) and after (ρ_c) the light curing, using the Archimedes' principle similar to what was described in PICK's study [16]. The mass (m) of uncured samples were measured on a precision balance and its volume (v) was measured with a pipette, than the initial density (ρ_u) was calculated:

$$\rho_u = m/v$$

After light curing for 60s (Bluephase G2, 1200 mW/cm²), the final mass of the samples were measured in air and water and then the final density (ρ_c) was calculated. The measurement was done immediately after curing and after 24h of dry storage at 37°C. The VS (vol%) was calculated by the following equation:

$$VS = (\rho_c - \rho_u / \rho_c) \times 100$$

where: ρ_c - the final density (cured), ρ_u - the initial density (uncured).

Statistical analyses

The data were analyzed by one-way ANOVA and post hoc Tukey's test. Statistical significance was established at $\alpha=0.05$ for all tests.

Results

The FS, EM, DC, UTS and DCU values are shown in Table 2. The Bis-GMA/TEGDMA resins presented higher DC compared to the resins containing IBOMA, except for Resin 1(60/0/40) ($p \leq 0.001$). FS and EM for the IBOMA alone, regardless of the concentration, showed lower values compared to the other groups ($p \leq 0.001$). UTS showed that in general, the resins containing only IBOMA as diluent monomer presented lower values, compared to the Bis-GMA/TEGDMA systems ($p \leq 0.001$). The DCU decreased with the presence of IBOMA, regardless the concentration ($p \leq 0.001$). The only exception occurred for Resin 3 (40/0/60), with the higher concentration of diluent monomer, which also showed lower DCU.

The VS (Table 3) was modulated by the inclusion of IBOMA in the resin formulations. Either IBOMA alone or IBOMA/TEGDMA showed immediate and 24h VS lower than the Bis-GMA/TEGDMA series ($p \leq 0.001$).

Table 2. Means (standard deviation) for the FS, EM, DC, UTS and DCU for the tested experimental resin blends.

Resins					
Bis/TEG/IBOMA	FS (MPa)	EM (GPa)	DC (%)	UTS (MPa)	DCU (mm)
R1 - 60/40/0	127.8 (20.8) A	1.70 (0.10) A	53.9 (5.3) B	68.2 (10.0) A	14.9 (0.3) A
R2 - 50/50/0	122.9 (15.9) A	1.59 (0.17) AB	61.2 (5.5) A	63.9 (6.6) AB	14.4 (0.7) A
R3 - 40/60/0	112.2 (28.1) A	1.62 (0.15) AB	61.9 (6.0) A	55.4 (10.5) B	11.2 (0.4) C
R4 - 60/0/40	78.0 (24.7) B	1.34 (0.28) B	55.2 (4.6) B	45.5 (4.9) C	10.8 (0.8) C
R5 - 50/0/50	80.2(16.5) B	1.32 (0.15) B	54.5 (4.5) B	42.6 (5.6) C	10.8 (0.4) C
R6 - 40/0/60	66.9 (15.9) B	1.35 (0.23) B	54.1 (0.9) B	39.4 (4.8) C	11.0 (0.6) C
R7 - 60/20/20	127.1 (23.3) A	1.73 (0.29) A	47.3 (5.4) C	56.3 (6.8) B	12.6 (0.8) B
R8 - 50/25/25	133.2 (22.4) A	1.77 (0.32) A	52.4 (6.6) B	45.9 (6.7) C	11.2 (1.0) C
R9 - 40/30/30	123.7 (15.4) A	1.54 (0.07) AB	52.5 (6.8) B	48.7 (9.3) B	10.1 (0.4) C

Same upper letter indicates no statistical difference among the values.

Table 3. Means (standard deviation) for the immediate volumetric shrinkage (Im VS) and 24h volumetric shrinkage (24h VS) for the experimental resin blends.

Resins		
Bis/IBOMA/TEG	ImVS (%)	24h VS (%)
R1 - 60/0/40	6.88 (0.30) BC	7.17 (0.36) BC
R2 - 50/0/50	7.13 (0.56) B	7.51 (0.23) AB
R3 - 40/0/60	8.11 (0.26) A	8.36 (0.27) A
R4 - 60/40/0	6.10 (0.53) CD	6.36 (0.37) CD
R5 - 50/50/0	4.45 (0.29) EF	4.03 (0.47) F
R6 - 40/60/0	3.64 (0.43) F	3.90 (0.97) F
R7 - 60/20/20	4.57 (0.51) E	5.06 (0.26) E
R8 - 50/25/25	6.26 (0.54) BCD	6.37 (0.57) CD
R9 - 40/30/30	5.52 (0.34) D	5.72 (0.24) DE

Same upper letter indicates no statistical difference among the values.

Discussion

The tested hypothesis was partially validated, since the IBOMA addition in the model dental resins allowed lower VS; however, the resins presented similar or reduced physical-mechanical properties compared to the Bis-GMA/TEGDMA series. It is well known that the VS is directly related to the concentration and DC of the carbon double-bonds [17]. Consequently, lower DC implies on lower VS and shrinkage stress [17,18]. In the present study, Bis-GMA/IBOMA showed lower VS, probably due to the lower DC values. Resin 4, which contained Bis-GMA/IBOMA (60/40 wt%), exhibited higher VS compared to the other resins with this co-monomer concentration (50 wt% and 60 wt%). The reason for this is maybe due to IBOMA, which is a lower shrinkage monomer, is reduced in the formulation and the base monomer is increased, although the DC is similar among the resins containing Bis-GMA/IBOMA. He *et al.* (2011), found that for a resin blend containing Bis-GMA/IBOMA 50/50wt%, the VS is around 4.05% and for Bis-GMA/TEGDMA 50/50 wt%, 8.79%, similar to the observed in this study.

The higher curing performance for the TEGDMA-based resins can be explained by the concentration of carbon double-bonds present in the resin system. Since IBOMA is a monomethacrylate, it presents only one carbon double-bond, when compared to the dimethacrylate TEGDMA, which contains two carbon double-bonds. As a consequence, the DC and VS were lower for the Bis-GMA/IBOMA when compared to the Bis-GMA/TEGDMA resin. For the resin 7, the high content of Bis-GMA and only 20 wt% of TEGDMA [13,14], may be the reason for only 47.3% of monomer conversion. The DCU showed that higher the content of IBOMA, lower the depth of cure. It is well explained by the lower DC achieved by the IBOMA resins. Also, the Bis-GMA/TEGDMA with 40 wt% of the diluent co-monomer showed lower DC, probably because they should be poorly polymerized and be softening during the scrapping away approach of the assay.

In clinical conditions, several factors could be determinant in the depth of cure of the composite resins, such as monomer type, inorganic filler size, photoinitiator concentration, light unit used and duration of irradiation. In this *in vitro* study, due to similarity in experimental conditions, the higher depth of curing in R1 and R2 could be directly tied to the difference in organic matrix nature. It was obvious that samples that

presented lower depth of cure would also present reduction DC, compromising several important already mentioned properties.

For this “scraping” test, a 20mm deep mold was used, differing from the ISO 4049:2009 [15] specifications, for the experimental matrix did not contain filler, and was more likely to present lower viscosity. Yet the ISO 4049:2009 [15] method was developed using a microfilled resin composite (Durafill, Kulzer & Co GmbH, Bad Homburg, West Germany). So the raise in depth was chosen to deliberately force the outcome of a difference in light scattering quality. For that matter, one theory that could explain the lower depth of cure in groups containing IBOMA along with the group Bis-GMA/TEGDMA (40/60%wt) regards their lower viscosity and relative light absorption and transmission quality. Usually, when the density of a medium increases its refractive index also increases. It is called ‘refraction of light’ the changing in its propagation medium, accompanied by variation in the propagation speed. The speed of light change in the medium is related to the change in composition of the sample, which changes the refractive index. It could be speculated that the groups containing TEGDMA, and mostly BisGMA in greater proportion, present higher refractive index. The higher the refractive index is, regardless of whether it increases during the polymerization phase of the monomers, the higher the light scattering and the lower the light transmission will be.

The photons have to perform successive collisions with the molecules within the resin bulk resulting in a delay, reducing its speed. Extended presence of larger amount of photons means increased availability of photons to participate in the reaction[20]. Theoretically, samples containing only IBOMA as diluent facilitate the passage of photons and thus have its polymerization hindered.

A simpler theory says that when the light first hits the surface, it would immediately interact with the photoinitiator system, causing the conversion of monomers. The premature polymerization of the surface layer of the sample would serve as a physical barrier to the passage of light. The top surface of the sample would receive the full power density of the curing light during the irradiation while the deeper parts, because of the barrier, would receive light of lower power density.

Either way, further investigations are required to explain the experimental matrices behavior towards light incidence, so that photopolymerization methods, photoinitiators ratios, or the need for accelerator addition can be assessed. A recent study [21] investigated depth of cure using numerous measurement techniques and the authors concluded that not only depth of cure measured similarly to the ISO 4049 method but also Vickers hardness profiles overestimated depth of cure.

As seen, the higher the IBOMA content, lower the VS. It may be occurred since the high IBOMA content, allow the polymer network to present less conversion of carbon double-bonds and consequently, lower VS. One important and positive fact observed in the results of the present work is that all the Bis-GMA/IBOMA resins (60/40, 50/50 and 40/60 wt%) showed similar DC as the Bis-GMA/TEGDMA resin (60/40 wt%), which is a common polymeric matrix in the direct resin materials; therefore, they present the same curing potential also showing reduced VS values, what would collaborate with the durability of the restorations [10,22].

The FS is an important property to predict the mechanical resistance of the resin restorations. The Bis-GMA/IBOMA resins showed lower FS compared to all other resins. Nevertheless, the elastic modulus increased with the TEGDMA presence, for all tested resins. It occurs due to the higher DC and high crosslink densities of the Bis-GMA/TEGDMA resins. The monofunctional structure of IBOMA compromises the polymer matrix because it tends to generate lower degree of conversion and lower cross-link density, showing lower EM and FS results for these resins[7,14]. Although the aliphatic cyclic structure of the monomethacrylate IBOMA hypothetically may have reinforced the resin blend, the double functional structure promoted by the inclusion of TEGDMA shows better results. Hence, when the TEGDMA was associated, the IBOMA presented similar values to TEGDMA resins blends.

The UTS is highly related to the cross-link density of the polymer, as well as to the monomer conversion and matrix composition [10]. In the present study, IBOMA alone promoted lower monomer conversion and may have led to low cross-link density and UTS. In this sense, the cohesive strength of the polymer backbone may be compromised in these

formulations. However, for the Bis-GMA/TEGDMA resins, the diluent co-monomer is proved to be a high cross-link monomer and with an increased cure potential [7,23,24].

Assessing the properties of resins containing IBOMA needs more investigation since when associated to TEGDMA, it showed some good results. Additional tests such as bond strength, marginal adaptation and shrinkage stress must be encouraged in order to achieve an interesting formulation for dental practice, in which the resin material have low shrinkage and good physical-mechanical properties. Also, further clinical investigations should be processed to predict the lifetime of the new monomer compositions for dental resins. Moreover, the inclusion of inorganic filler particles is important for a global evaluation of the performance of IBOMA as reactive diluent co-monomer in dental resins, especially the resin composites.

Conclusion

Within the limitations of the present study, it can be concluded that the IBOMA used alone or in association with TEGDMA in the experimental resin blends showed lower volumetric shrinkage, when compared to TEGDMA alone as copolymer. However, resin blends with IBOMA alone or in association with TEGDMA, led to lower physical-mechanical properties values, such as degree of conversion, ultimate tensile strength and depth of cure compared to the Bis-GMA/TEGDMA systems and similar results of flexural strength and elastic modulus.

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Capítulo 2

Physical properties of experimental resins blends containing the monomethacrylate IBOMA as diluent co-monomer

Abstract

Objective: The aim of this study was to evaluate the Knoop hardness (KHN), crosslink density (CLD) and water sorption (WS) of experimental resins blends in function of the copolymer content and concentration.

Materials and methods: A blend of Bis-GMA as base monomer was formulated with different concentrations of diluent, such as TEGDMA(60/40, 50/50 e 40/60wt%), IBOMA (60/40, 50/50 and 40/60wt%), or TEGDMA/IBOMA (60/20/20, 50/25/25 and 40/30/30 wt%). The Camphoroquinone 0.5wt% / 2-(N,N-Dimethylamino)ethyl methacrylate 1wt% (CQ/DMAEMA) was used as the photoinitiator system and inhibitor butylated hydroxytoluene 0.1 wt% (BHT). The KHN was measured at top surface using an indenter. Polymer softening in ethanol was used as an indirect method for assessing the degree of crosslinking. For WS, the volume of the samples was calculated. They were transferred to desiccators until a constant and were subsequently immersed in distilled water for 7 days and then weighed again until no alteration in mass was detected (m_1). Samples were reconditioned to constant mass in desiccators (m_2). WS was determined using the equations $m_2 - m_3/V$ and $m_1 - m_3/V$, respectively. Data were submitted to ANOVA and Tukey's test ($p \leq 0.05$).

Results: The resins containing Bis-GMA/TEGDMA 60/40% and Bis-GMA/TEGDMA/IBOMA 60/20/20 wt% showed higher KHN values. The IBOMA groups showed lower CLD, while TEGDMA groups had higher values of CLD. The highest values of WS were presented by the group where TEGDMA was the diluent monomer, and for the WSOL all groups showed no significant differences among themselves.

Conclusion: IBOMA in association with TEGDMA showed improvements in their physical-mechanical properties, however, showed a decrease in CLD and KHN compared to TEGDMA alone, when used as diluent co-monomer Bis-GMA dental resins.

Keywords: methacrylate monomers, dental resin, curing performance.

Introduction

Composite resins are highly successful restorative materials in dentistry. However, there are factors that create concerns due to deleterious processes caused by the stress generated during the polymerization reaction [1]. Lost dental structures are being replaced by these dental restorative materials, due to the need for aesthetic or loss of function of the tooth, since they have presented good results on modern dental research in biomaterials.

Dental composites are derived from methacrylate networks and the base monomer most currently used in these composites is bisphenol glycidyl methacrylate (Bis-GMA), known to be somewhat volatile, to present low diffusivity between tissues and low shrinkage [2]. The high viscosity of Bis-GMA monomers requires the addition of low molecular weight monomer in order to obtain a suitable viscosity and improve mobility of the monomers during the polymerization reaction, increasing the degree of conversion and, in addition, facilitating the incorporation of inorganic particle fillers [3,4].

Due to its low viscosity and favoring to an increase in the degree of conversion, triethyleneglycol dimethacrylate (TEGDMA) is a diluent monomer most commonly added to the base monomer [5,6]; however, it exhibits high volumetric shrinkage [7]. The addition of TEGDMA in greater proportion increases the polymerization shrinkage and water sorption of the matrix [8]. Several alternative diluent monomers have been tested in the attempt to a partial or total substitution of TEGDMA and consequent reduction of these drawbacks [9].

The isobornyl methacrylate (IBOMA) is a monomethacrylate that presents low viscosity and polymerization shrinkage. Studies show its use in the synthesis of nanogels for resin matrix aiming to reduce shrinkage and polymerization stress [10,11]. IBOMA is also used as a co-monomer thinner for the composite matrix due to its low viscosity, low polymerization shrinkage and high hydrophobicity [12]. Besides, this co-monomer presents low water absorption, which can increase the durability of the polymer due to the resistance to degradation, in the oral environment [13].

The phenomena of sorption and solubility may be precursors to a variety of chemical and physical processes that promote biological concerns as well as having deleterious effects on the structure and function of resin matrixes [14]. Polymer structure

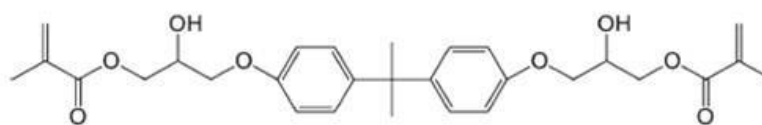
quality such as the degree of conversion and crosslink density resulting from the photo-activation mode may lead to differences in sorption and solubility [15,16].

Based on this, the aim of this study was to evaluate the physical properties of experimental resin blends using Bis-GMA as monomer base, IBOMA as diluent monomer alone or in combination with TEGDMA, in different proportions. The hypothesis tested in this study is that the addition of alternative diluent monomer (IBOMA) may minimize the water absorption without compromising the Knoop hardness and crosslink density of these experimental resins.

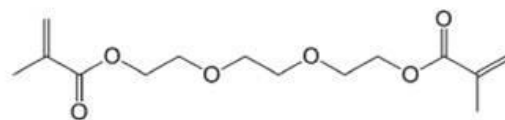
Materials and methods

Resin preparation

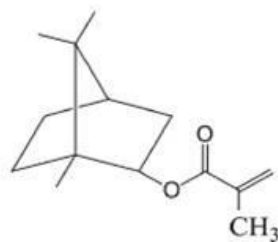
Nine experimental resin formulations were tested in this study. The resin matrix consisted on bisphenol glycidyl methacrylate (Bis-GMA - Sigma-Aldrich Inc, St Louis, MO, USA) as base monomer and two diluent reactive co-monomers: triethyleneglycol dimethacrylate (TEGDMA - Sigma-Aldrich Inc, St Louis, MO, USA) and isobornyl methacrylate (IBOMA - Sigma-Aldrich Inc, St Louis, MO, USA). The structure of these molecules of monomers is showed in the Figure 1. The diluent co-monomers were mixed with the base monomers in nine different proportions, as described in Table 1. The photoinitiator system was composed of camphorquinone (CQ – 0.5 wt%, Sigma-Aldrich Inc, St Louis, MO, USA and 2-(dimethylamino)ethyl methacrylate (DMAEMA – 1 wt%, Sigma-Aldrich Inc, St Louis, MO, USA). Also, the inhibitor BHT (butylated hydroxytoluene, Sigma-Aldrich Inc, St Louis, MO, USA) was added to the organic matrix in a concentration of 0.1 wt% to avoid spontaneous polymerization of the monomers.



Bis-GMA



TEGDMA



IBOMA

Figure 1. Bis-GMA, TEGDMA and IBOMA molecules.

Table 1. Composition of the experimental resin blends.

Resin	Bis-GMA%	TEGDMA%	IBOMA%
Resin 1 - 60/40/0	60	40	-
Resin 2 - 50/50/0	50	50	-
Resin 3 - 40/60/0	40	60	-
Resin 4 - 60/0/40	60	-	40
Resin 5 - 50/0/50	50	-	50
Resin 6 - 40/0/60	40	-	60
Resin 7 - 60/20/20	60	20	20
Resin 8 - 50/25/25	50	25	25
Resin 9 - 40/30/30	40	30	30

Knoop Hardness

To the KHN, circular samples (n=10) were prepared (2mm thickness x 5mm diameter) and light cured by a LED curing unit (Bluephase G2, 1200 mW/cm²), during 60s. The total energy dose was standardized at 72J. Then, the samples were dry stored at 37 °C for 24h in light-proof containers. Thereafter, the top surface was wet-polished with 1,200-grit SiC paper to obtain a planar surface. Knoop hardness measurements were taken using an indenter (HMV-2, Shimadzu, Tokyo, Japan), under a load of 490 N, for 15 s. Five readings were performed for each specimen. The KHN (in kilogram–force per square millimeter) was recorded as the average of the five indentations. Data were submitted to one-way ANOVA followed by Tukey's test (p=0.05).

Crosslink Density

After completion of the Knoop Hardness test, the same samples (n=10) were used in the CLD test. The samples were immersed in 100% ethanol, for 24h, to evaluate indirectly the CLD and the elution of monomers, by KHN mean. The Knoop hardness measurements were taken on top surface using an indenter (HMV-2, Shimadzu, Tokyo, Japan), under a load of 490 N (equivalent to 50 gf) for 15s. Five readings were performed for each sample.

The KHN was recorded as the average of the five indentations. Data were submitted to one-way ANOVA followed by Tukey's test ($p \leq 0.05$). Additionally, the percentage decrease of KHN was calculated.

Water sorption

This study was performed in compliance with ISO 4049:2009 [17] standard specifications (except for the specimen dimensions and curing protocol) as follows. To test the WS, circular samples (2mm thickness x 5mm diameter) were prepared ($n=5$) and light cured by LED curing unit (Bluephase G2, 1200 mW/cm^2), during 60s. The total energy dose was standardized at 72J. The disks were stored in desiccators containing silica gel at 37°C for 24 hours and the thickness (four measurements at four equidistant points on the circumference) and diameter (two measurements at the right angles) of each specimen were measured using a digital electronic caliper (Mitutoyo Corporation, Tokyo, Japan). The mean values were used to calculate the volume (V) of each specimen (in mm^3). Thereafter, the samples were stored with distilled water at 37°C for 7 days. The volume of water immersion was 6ml per specimen. Samples were weighted daily, after being carefully wiped with an absorbent paper. When constant weight was obtained (two days of no weight change), this value was recorded as m_1 . After this weighing, the samples were returned to the desiccator. The entire mass reconditioning cycle was repeated and the constant mass (two days of no weight change) was recorded as m_2 .

The WS values, in micrograms per cubic millimeters, were calculated using the following equations:

$$\text{WS} = (m_1 - m_2) / V$$

Statistical analyses

The data were analyzed by one-way ANOVA and post hoc Tukey's test ($p \leq 0.05$).

Results

The KHN and CLD values are shown in Table 2 and Figure 2. The resin 1 (Bis-GMA/TEGDMA/IBOMA 60/40/0wt%) and resin 7 (Bis-GMA/TEGDMA/IBOMA 60/20/20wt%) showed the highest values of KHN, and resin 6 (Bis-

GMA/TEGDMA/IBOMA 40/0/60 wt%) showed the lowest values. The IBOMA groups (resins 4-6) had the lowest means for CLD, while the TEGDMA groups (resins 1-3) showed higher value CLD. The resin groups where both diluents monomers were present (resins 7-9) had intermediate values. The hardness decrease for TEGDMA groups ranged 45.65-54.92%; for IBOMA groups ranged 74.05-81.02% and for TEGDMA-IBOMA groups ranged 63.66-69.39%.

The WS data are listed in Table 3. The Bis-GMA/ TEGDMA resins showed the highest values of WS.

Table 2. Means and standard deviation of the Knoop hardness, cross-link density and hardness decrease for the experimental resins.

Resins	Hardness Knoop Inical (KH)	Hardness Knoop after ethanol 24h (KH)	Hardness Knoop decrease (%)
R1 - 60/40/0	35.5 (4.8) A	16.0 (2.0) A	54.92
R2 - 50/50/0	31.1 (4.1) AB	16.9 (1.8) A	45.65
R3 - 40/60/0	31.4 (5.7) AB	16.8 (2.0) A	46.49
R4 - 60/0/40	31.1 (3.8) AB	5.9 (0.6) C	81.02
R5 - 50/0/50	26.7 (3.7) BC	6.6 (0.9) C	75.28
R6 - 40/0/60	23.9 (2.8) C	6.2 (0.6) C	74.05
R7 - 60/20/20	33.0 (2.2) A	10.1 (0.6) B	69.39
R8 - 50/25/25	31.1 (3.3) AB	10.4 (0.9) B	66.55
R9 - 40/30/30	32.2 (4.5) AB	11.7 (0.9) B	63.66

Same upper letter indicates no statistical difference among the values.

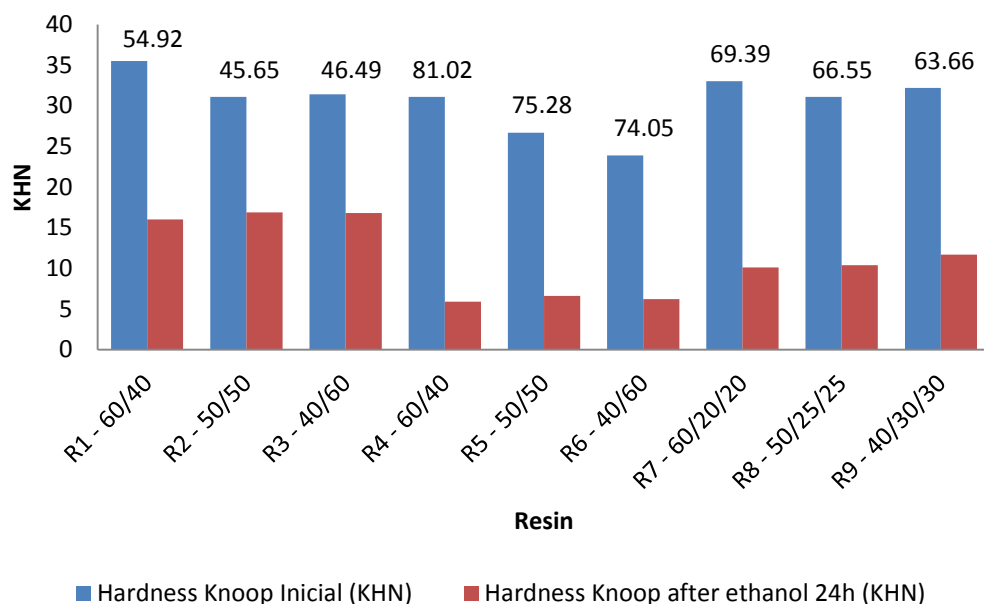


Fig.2 – Graphic presenting the cross-link density survey with the Knoop hardness before and after immersion in absolute ethanol. Knoop hardness reduction (%) presented above the columns.

Table 3. Means and standard deviation of the water sorption for the tested experimental resins.

Resins	Water sorption
R1 - 60/40/0	35.5 (5.1) A
R2 - 50/50/0	41.4 (8.5) A
R3 - 40/60/0	48.4 (4.2) A
R4 - 60/0/40	22.9 (6.2) BC
R5 - 50/0/50	15.3 (3.2) C
R6 - 40/0/60	15.4 (4.7) C
R7 - 60/20/20	26.2 (5.3) BC
R8 - 50/25/25	29.2 (4.2) B
R9 - 40/30/30	24.6 (4.8) BC
p = 0.1559	

Same upper letter indicates no statistical difference among the values.

Discussion

The hypothesis was partially validated, since experimental resins containing IBOMA as monomer diluent, presented the lower KHN and CLD values; however, it also showed the lower WS values when compared with Bis-GMA/TEGDMA resins. WS in resin-based materials is a diffusion-controlled process and occurs mainly in the resin matrix [18]. In this study, it was observed high values of WS when the diluent monomer TEGDMA was present in the resin matrix. According to SIDERIDOU (2003), higher TEGDMA content in the matrix is responsible for increasing the WS of the composites. The WS of the copolymer is influenced by its hydrophilicity [10] and crosslink density [21].

When higher crosslink density occurs, the hole-free volume between polymer chains is reduced decreasing polymer plasticizing as well. ASMUSSEN and PEUTZFELD (2001) verified the influence of the composition on crosslink density of experimental composites containing different variations of TEGDMA/Bis-GMA, using hardness test before and after immersion in absolute ethanol. They observed that the variation in the composite composition influenced the crosslink density. Also, a decrease in the hydrophilicity and an increase in the crosslink density of a copolymer could reduce the water sorption of the matrix [12]. However, the crosslink densities of the Bis-GMA/IBOMA and Bis-GMA/IBOMA/TEGDMA were lower than that of the Bis-GMA/TEGDMA. The IBOMA is considered to be more hydrophobic than TEGDMA, which would lead to lower values of WS, but on the other hand, presents low ability to form crosslink among the polymer chains. CUI *et al.* (2007) verified that as IBOMA is a monomethacrylate, it presents low polymerization degree and, therefore, has fewer sites for crosslink in the polymer chain formation.

Knoop Hardness test is widely employed as an indirect method of determining the degree of conversion. It is reasonable to say that a low DC would increase the hole-free between polymer chains, thereby favoring the plasticizing effect of ethanol, although this effect mostly depends on the linearity of the polymer structure. In this study, the resins with IBOMA alone had lower KHN when compared with the TEGDMA groups. This fact can be explained by the fact that monomethacrylates as IBOMA tend to form linear polymers

when polymerized alone or in resin blends, unlike what happens with the TEGDMA, which is known as conventional crosslinkers in polymers [13].

IBOMA associated to TEGDMA showed good or intermediate physico-mechanical properties. Also, it could be an alternative to improve the organic matrix of the composites, since showed similar KHN values when compared to TEGDMA groups. However, further investigations should be conducted in order to clarify not only the durability, marginal adaptation, bond strength of this type of resin blend, but to get an interesting formulation for the dental practice as well. Another important factor is the inclusion of inorganic filler particles for evaluating the performance of IBOMA as reactive diluent monomer in dental resins.

Conclusion

Within the limitations of this study, the results suggest that experimental resins containing only IBOMA as diluent are significantly more susceptible to softening in ethanol than resin groups containing only TEGDMA, presenting lower KHN before and after this reaction. On the other hand, this same group shows lower WS values when compared with TEGDMA groups. The association of both diluents presented similar KHN values when compared to TEGDMA groups, suggesting that the IBOMA/TEGDMA association as diluents may be an alternative for maintaining good mechanical properties while diminishing water sorption.

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CONSIDERAÇÕES GERAIS

A busca pela redução da contração de polimerização é ainda um dos principais objetivos dos fabricantes de resinas compostas odontológicas, sendo ela, a responsável por grande parte dos problemas de longevidade das restaurações. Compósitos odontológicos baseados em siloranos de baixa contração foram introduzidos, e têm mostrado resultados promissores. Uma alternativa simples para o aprimoramento dos compósitos odontológicos é através da introdução de nanogéis que são nanopartículas poliméricas reticuladas sintetizadas, compostas pelos monômeros IBOMA e UDMA.

Nessa busca por melhores propriedades físico-mecânicas e de acordo com trabalhos mostrados na literatura, este trabalho buscou mais uma alternativa para os compósitos odontológicos com a incorporação do IBOMA como monômero diluente para diminuir a contração e a tensão de polimerização de matrizes de resinas compostas, sem prejuízo às outras propriedades físico-mecânicas. Matrizes resinosas contendo o monômero diluente IBOMA sozinho ou associado ao TEGDMA apresentaram menores valores de contração volumétrica. Importante e positivo resultado observado no presente trabalho foi o fato de todas as resinas Bis-GMA/IBOMA (60/40, 50/50 e 40/60 wt%) apresentarem grau de conversão semelhante ao da resina Bis-GMA/TEGDMA (60/40 % em peso) que é uma matriz polimérica comum nos compósitos atuais, ou seja, apresenta o mesmo potencial de polimerização, com valores reduzidos de sorção em água, colaborando para a durabilidade do material.

Em geral, as resinas cujo monômero diluente era exclusivamente o IBOMA, apresentaram valores mais baixos de resistência máxima à tração, quando comparadas às resinas convencionais Bis-GMA/TEGDMA. Além disso, quanto maior a quantidade de IBOMA na resina, menor a profundidade de polimerização. No entanto, as resinas contendo a associação de IBOMA e TEGDMA, apresentaram resistência à flexão, módulo de elasticidade, grau de conversão e absorção de água similares (ou até superiores em alguns casos) às resinas contendo somente TEGDMA. Isso permite que o IBOMA seja considerado um monômero diluente com potencial para substituir parcialmente o TEGDMA em formulações de matrizes para compósitos dentais.

No entanto, investigações adicionais devem ser conduzidas a fim de esclarecer não só a durabilidade *in vitro* de matrizes contendo esse monômero, mas também a longevidade

clínica das restaurações, analisando adaptação marginal, resistência de união, buscando sempre aprimorar as propriedades físicas e mecânicas. Talvez, o próximo passo seja a incorporação de partículas de carga inorgânicas nessas matrizes monoméricas a fim de se avaliar as mesmas propriedades dentro deste novo contexto.

CONCLUSÃO

Nas condições deste estudo, pode-se concluir que o monômero diluente IBOMA pode ser uma alternativa na formulação de matrizes para resinas compostas dentais, já que as misturas contendo somente IBOMA como diluente ou, principalmente, em associação com o TEGDMA, apresentaram algumas propriedades físico-mecânicas superiores ou similares em comparação às misturas contendo somente TEGDMA.

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APÊNDICE

Pranchas de Figuras

Figura 1 - Ilustrações dos materiais utilizados.

A. IBOMA - Sigma-Aldrich Inc, St Louis, MO, USA

B. Bis-GMA - Sigma-Aldrich Inc, St Louis, MO, USA

C. TEGDMA - Sigma-Aldrich Inc, St Louis, MO, USA

D. BHT - Sigma-Aldrich Inc, St Louis, MO, USA

E. DMAEMA - Sigma-Aldrich Inc, St Louis, MO, USA

F. Camphorquinone - Sigma-Aldrich Inc, St Louis, MO, USA

Figura 1

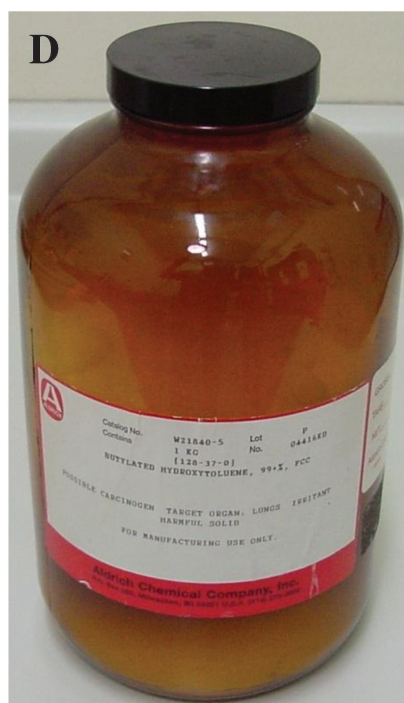
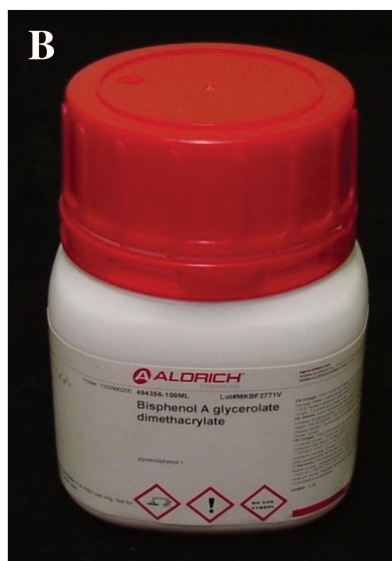


Figura 2 - Confecção (padrão) das amostras em formato de discos

1. Pera, béquer e pipeta
2. Peso do béquer tarado na balança para obtenção da massa da resina
3. Obtenção do volume por meio da pipeta, massa (peso) e posterior densidade
4. Tira de poliéster, lâmina de vidro e matriz de silicone
5. Inserção da resina na matriz com auxílio de gotejador
6. Matriz preenchida por resina
7. Posicionamento da tira de poliéster sobre a resina
8. Tira de poliéster posicionada
9. Lâmina de vidro colocada sobre a tira de poliéster
10. Led Bluphase G2 posicionado
11. Fotoativação por 60s
12. Espécime pronta
13. Dispositivo adaptado à balança, para obtenção da densidade dos espécimes após polimerização.

Figura 2

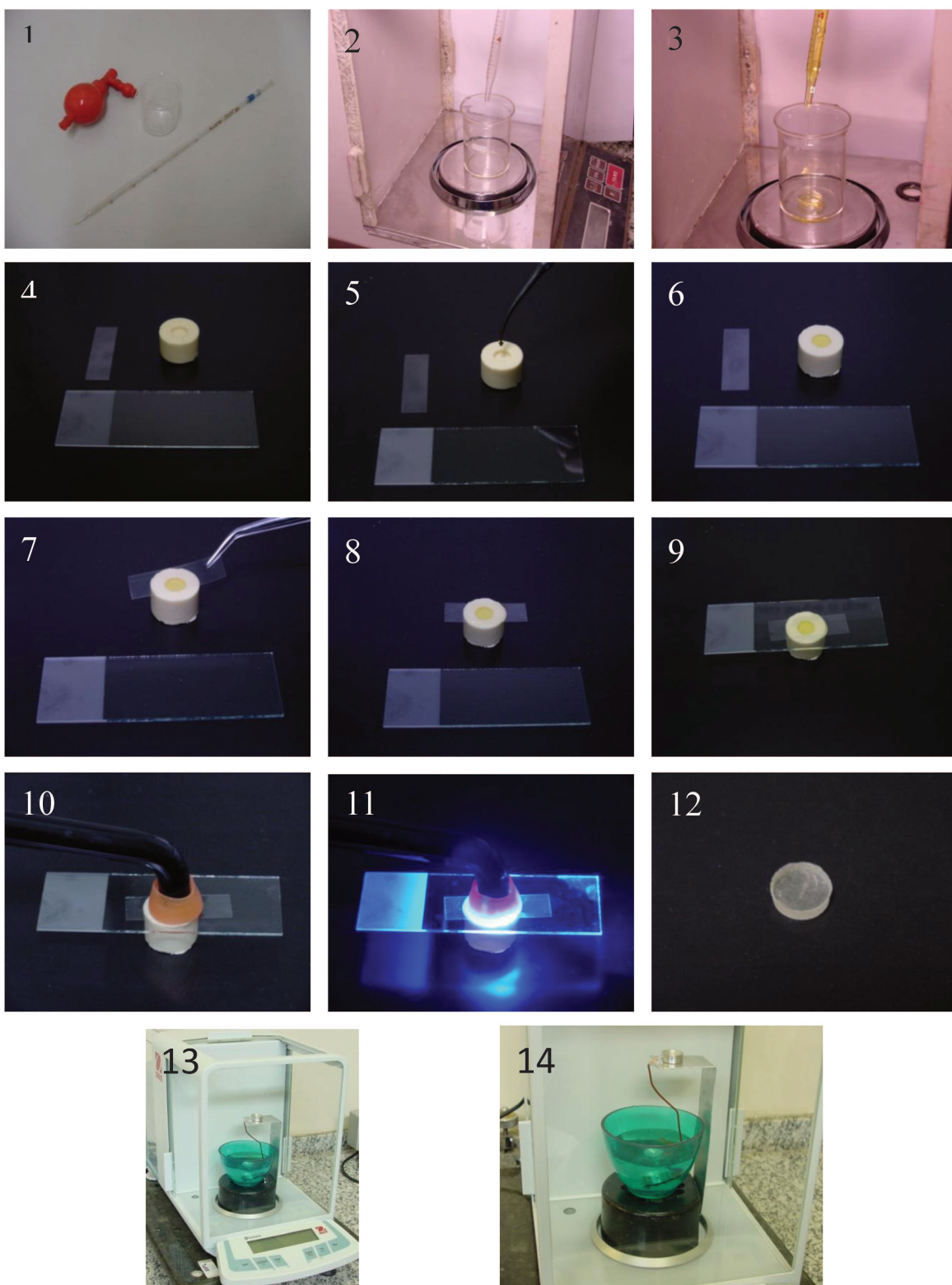


Figura 3 – Confeção dos espécimes em formato de ampulheta

1. Tira de poliéster, lâmina de vidro e matriz de silicone
2. Inserção da resina na matriz com auxílio de gotejador
3. Matriz preenchida por resina
4. Posicionamento da tira de poliéster sobre a resina
5. Tira de poliéster posicionada
6. Lâmina de vidro colocada sobre a tira de poliéster
7. Led Bluphase G2 posicionado
8. Fotoativação por 60s
9. Espécime pronta
10. Fixação da amostra em dispositivo próprio para a máquina de ensaio
11. EZ-Test, Shimadzu, Kyoto, Japan
12. Teste sendo realizado
13. Espécime após teste de microtração

Figura 3

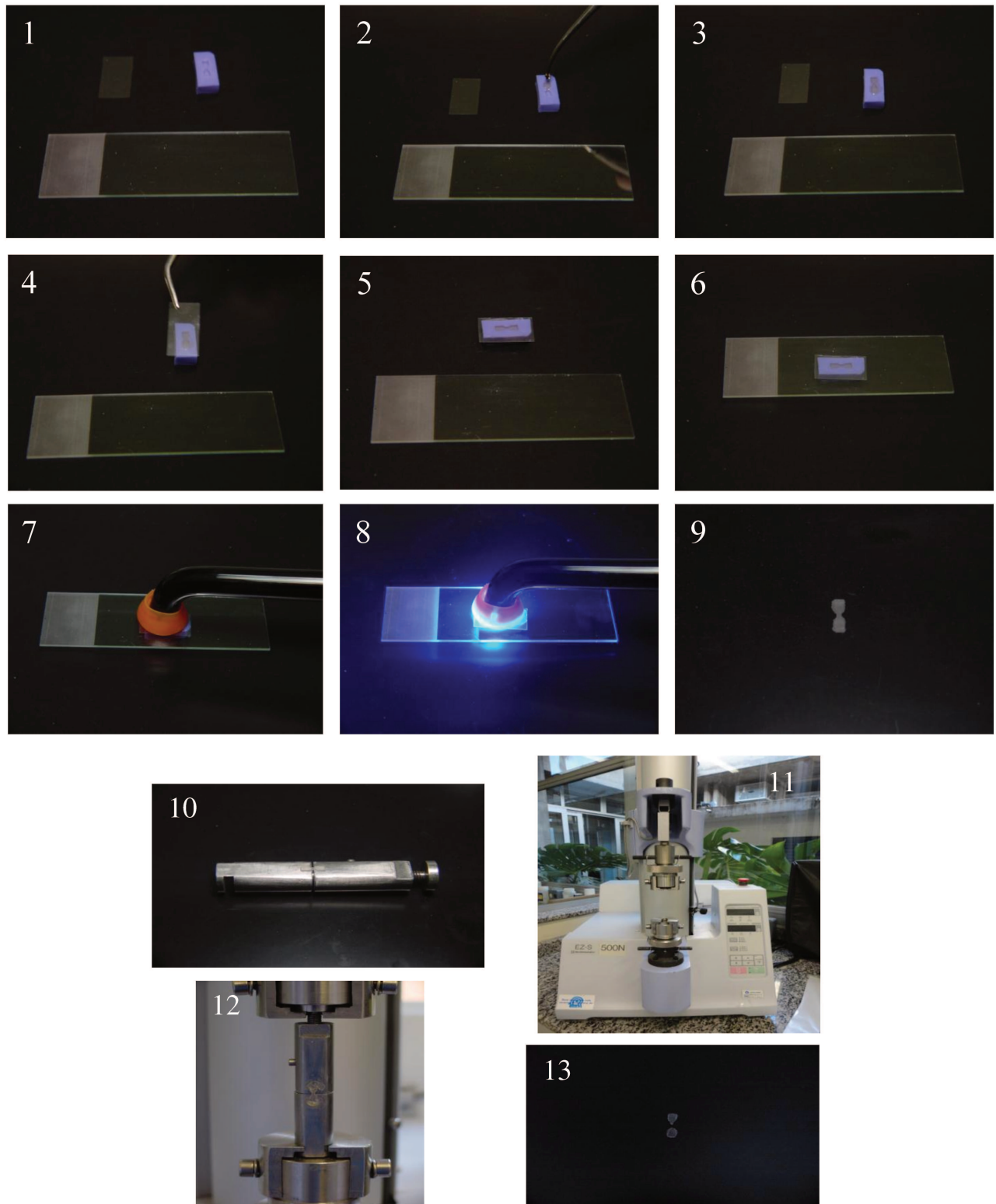
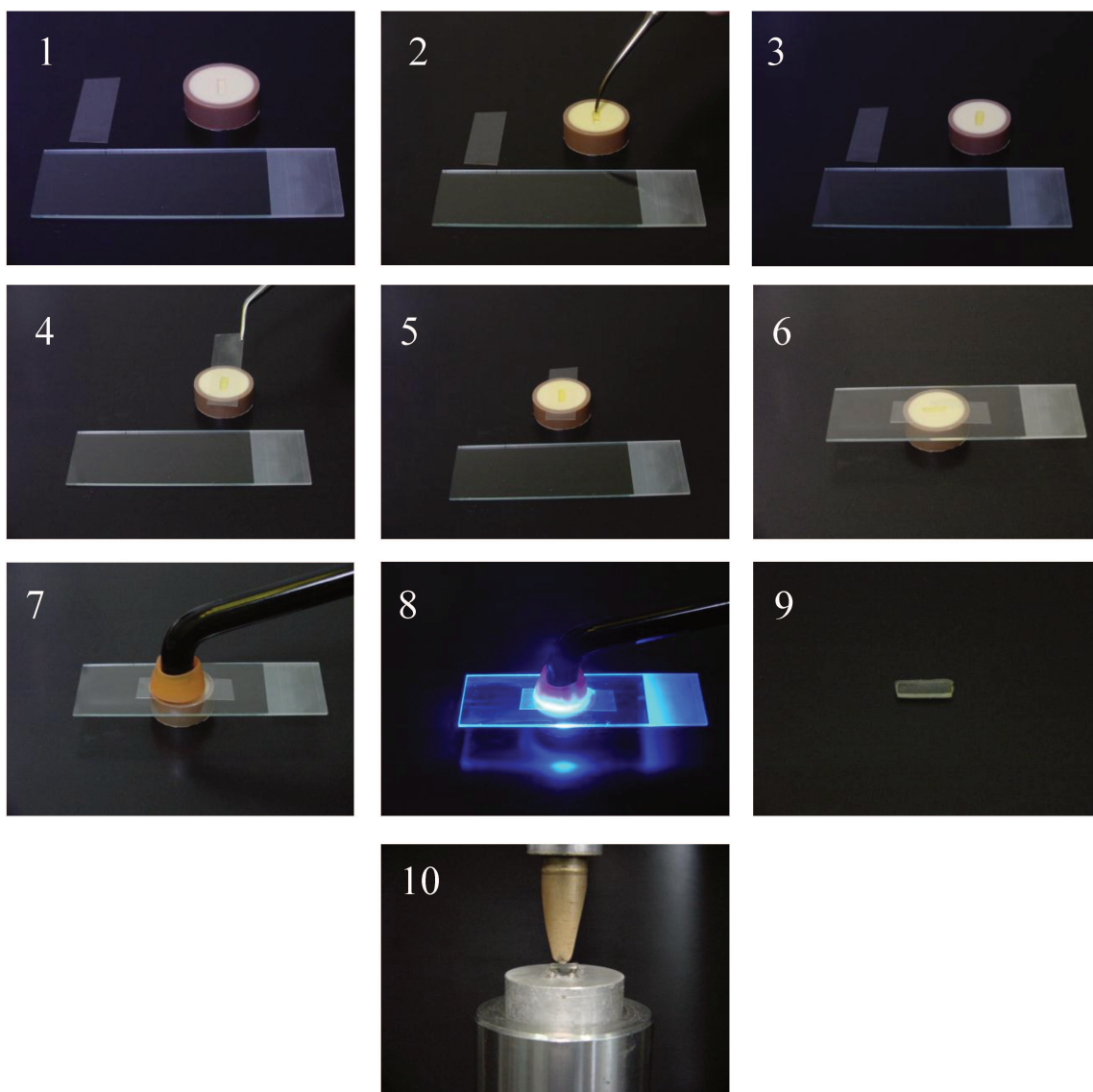


Figura 4 – Confeção dos espécimes em formato de barra

1. Tira de poliéster, lâmina de vidro e matriz de silicone
2. Inserção da resina na matriz com auxílio de gotejador
3. Matriz preenchida por resina
4. Adaptação da tira de poliéster sobre a resina
5. Tira de poliéster adaptada
6. Lâmina de vidro posta sobre a tira de poliéster
7. Led Bluphase G2 posicionado
8. Fotoativação por 60s
9. Espécime pronta
10. Realização do teste de resistência à flexão

Figura 4



ANEXO

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