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Solvent Degradation and Polymerization Shrinkage Reduction of Resin Composites Using Isobornyl Methacrylate

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The aim of this study was to use the isobornyl methacrylate (IBOMA) as a combining or substituent diluent monomer in the resin matrix of dental resin composites. Thus, the resin matrix was formulated with 60 wt% of BisGMA and 40 wt% of diluent monomers. TEGDMA as the only diluent monomer was used as control with 40 wt%, while total substitution of TEGDMA was done with 40 wt% of IBOMA. The combination of IBOMA and TEGDMA was done with 20 wt% of each monomer. To the resin matrix, 65 wt% of filler particles was added. Degree of conversion (DC) using FT-IR, flexural strength (FS), flexural modulus (FM), polymerization shrinkage by gap formation (GF), Knoop hardness (KH) and solvent degradation (SD) were evaluated. Data were analyzed using ANOVA and Tukey's test ($\alpha=0.05$; $\beta=0.2$). The results showed that reducing or substituting TEGDMA using IBOMA did not affect the DC (0.085), FS ($p=0.886$) or FM ($p=0.414$). Also, when IBOMA was used, lower GF was found in comparison to the control containing only TEGDMA as the diluent monomer ($p=0.032$). However, even though all composites presented reduction in KH during the SD test, the combination of IBOMA and TEGDMA showed similar reduction in KHN in comparison to the control group ($p=0.001$), while the total substitution of TEGDMA with IBOMA decreased KHN after SD ($p=0.041$). Thus, the combination of IBOMA and TEGDMA seem to reduce SD and GF without affecting the properties of resin composites.

Key Words: dental materials, dental resin, methacrylate, monomers.

Introduction

The resin composites are considered versatile and reliable for both direct and indirect dental restorations and due to their physical and esthetic properties, they have become one of the most used biomaterials in the restorative dentistry field (1). Regarding the basic composition of these materials, the dental composites consists in a high content of barium glass or silica filler particles as the disperse phase associated with high molecular weight monomers, such as the bisphenol-A glycidyl methacrylate (BisGMA) combined with one or more low-viscosity diluent monomers, such as the triethyleneglycol dimethacrylate (TEGDMA) as the continuous phase (2).

High molecular weight monomers generally exhibit low mobility that contributes to lower contraction but reduces the final resin composite degree of conversion (3) and increase the viscosity of the restorative material. The low viscosity diluent monomers are added to reduce the overall resin matrix viscosity, providing good handling properties and increase degree of conversion (4). However, the addition of diluents such as TEGDMA can increase the degradation of the material overtime, as well as the cytotoxic potential

of the material, because the unpolymerized molecules of this monomer diluent undergo saponification by enzymatic hydrolysis that breaks down the molecule of TEGDMA into two molecules of methacrylic acid. Moreover, due to its high lipophilicity, these molecules can penetrate rapidly into the membrane of human cells, and alter cells function (5,6). However, one of the major reasons for hydrolytic degradation of composites containing high amount of TEGDMA is that the TEGDMA molecule has two oxygen atoms in the central molecule backbone and each oxygen atom has 2 free electrons in their valence shell, which makes according to the Fick's first law the TEGDMA monomer more susceptible to water diffusion than other kinds of monomers. Thus, in order to develop materials with less degradation and less cytotoxic potential without affecting its chemical and mechanical properties, alternative diluents to TEGDMA have been proposed (7).

Isobornyl methacrylate (IBOMA) is a monomethacrylate monomer with low viscosity that has been studied as diluent in coatings resins and for polymer purposes (8,9), and showed lower water absorption and greater resistance to degradation. These particular properties characteristics

may increase the durability of the polymer in the oral medium. The molecular weight of IBOMA is similar to the TEGDMA, however the main difference is regarding the hydrolytic degradation, especially because IBOMA during the enzymatic hydrolysis process leads to the formation of only one molecule of methacrylic acid, showing lower cytotoxicity than TEGDMA that can lead to two methacrylic acid molecules originated from the enzymatic hydrolysis process (1). Additionally, IBOMA has a unique behavior of being less susceptible to water diffusion water than TEGDMA which makes polymer formed by IBOMA more water stable. Nonetheless, it is important to bear in mind that despite the fact that IBOMA is a monomethacrylate such as HEMA, the HEMA behavior in respect to water sorption and degradation is completely different from the IBOMA. The HEMA reduces the polymer crosslink density, which increases the leachability of non-reacted species, and that HEMA has a smaller molecule with higher potential to reach the pulp by dentin tubules. However, differently from HEMA, IBOMA is a heterocyclic molecule that does not have the any hydrophilic functional group, and according to the Fick's first law, the IBOMA is less susceptible to water diffusion due to the absence of hydroxyl functional group (-OH) that follows the diffusion laws and could lead to excessive polymer water uptake (7).

However, although monomethacrylates, such as IBOMA, might increase the monomeric conversion (10) and reduce the cytotoxic potential (1) of resin materials, the results of the mechanical properties can be significantly reduced in comparison to the dimethacrylates (10,11). This would occur due to the fact that monomethacrylate monomers, such as the IBOMA, do not contribute to polymer crosslinking as dimethacrylate monomers do (10,11). However, combining dimethacrylate and monomethacrylate monomers might be the key for not necessarily affecting crosslinking but delaying vitrification, and possibly reducing shrinkage as well. Thus, the aim of this study was to reduce TEGDMA concentration using IBOMA as a combining or substituent diluent monomer in the resin matrix of dental resin composites. The tested hypotheses were if: 1- reducing TEGDMA concentration using IBOMA as a combining diluent monomer will affect the properties of resin composite formulations; 2- substituting TEGDMA using IBOMA as the only diluent monomer will affect the properties of resin composite formulations.

Material and Methods

Resin Composite Formulations

Three experimental resin composite formulations were mechanically blended using a centrifugal mixing device (SpeedMixer, DAC 150.1 FVZ- K, Hauschild Engineering, Hamm, North Rhine-Westphalia, Germany). As described

in Table 1, the organic resin matrix of all formulations consisted on 60 wt% of Bis-GMA (Sigma-Aldrich Inc., St Louis, MO, USA) combined with 40 wt% of diluent monomer(s). The IBOMA (Sigma-Aldrich Inc.) was used as a diluent monomer for Bis-GMA, alone or in combination with TEGDMA (Sigma-Aldrich Inc.) (1:1). TEGDMA alone as the only diluent monomer was used as control. The chemical structure of each monomer is illustrated in Figure 1. To the resin matrix, 65 wt% of filler particles was added, in which, 13 wt% was 0.05 μm fumed silica (Aerosil OX50, Nippon Aerosil Co. Ltd., Yokkaichi, Tokyo, Japan) and 52 wt% was 0.7 μm BaBSiO₂ glass (Esstech Inc., Essington, PA, USA). A 1:4 filler ratio of silica and glass was used in order to assign proper handling properties to the composites. The photo-initiator system used in all formulations was 0.5 wt% of camphorquinone (Sigma-Aldrich Inc.) combined with 1 wt% of 2-(dimethylamino)ethyl methacrylate, DMAEMA (Sigma-Aldrich Inc.).

Degree of Conversion (DC)

For DC analysis, bar-shaped samples (7 mm in length x 2 mm in width x 1 mm in thickness) (n=10) were produced from molds under the same photo-activation protocol using a LED curing unit (Bluephase G2, Ivoclar Vivadent AG, Schaan, Liechtenstein) for 20 s of exposure. The irradiance of the LED curing unit was 1200 mW/cm², and the total energy exposure was 24 J/cm². After photo-activation, the samples were removed from the molds and dry stored in light-proof containers at 37 °C for 24 h. After this period, the spectra were collected on the top surface of each specimen using Fourier transformed infrared spectroscopy in conjunction with attenuated total reflectance (Spectrum 100, PerkinElmer, Shelton, CA, USA). For non-polymerized spectra, each experimental composite formulation was sandwiched between two glass plates with same thickness (1 mm).

The absorption spectra of non-polymerized and polymerized experimental composite resins were obtained from the region between 4000 and 650 cm⁻¹, with 32 scans at 4 cm⁻¹. The 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

Table 1. Composition of the experimental resin matrices used.

Diluent monomer ratio (TEGDMA:IBOMA)	Bis-GMA wt%	TEGDMA wt%	IBOMA wt%
1:0	60	40	0
0:1	60	0	40
1:1	60	20	20

component (located at 1608 cm^{-1} ; reference peak) were obtained. The DC (%) was calculated using the following equation:

$$\text{DC}(\%) = 100 \times [1 - (R_{\text{polymerized}} / R_{\text{nonpolymerized}})],$$

where, R represents the ratio between the absorbance peak at 1638 cm^{-1} and 1608 cm^{-1} .

Flexural Strength (FS) and Flexural Modulus (FM)

After the DC analysis, the same bar-shaped samples ($n=10$) were used for the three-point bending flexural test. The FS test was performed according to a new method rather than the ISO 4049:2009 (12), in which the specimen's dimensions were $7\text{ mm} \times 2\text{ mm} \times 2\text{ mm}$ (13) and the specimens were kept immersed in artificial saliva. This method increases significant differences in flexural strength in comparison to the ISO method, as well as allowing prediction of mechanical performance under more clinically realistic conditions (13). The three-point bending test was performed in a universal testing machine (Instron, Canton, MA, USA) with span between supports of 5 mm and 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: $\text{FS} = 3FL / (2BH^2)$, where: F was the maximum load (N) exerted on the samples; L was the distance (mm) between the supports; B was the width (mm) of the samples; and, H was the height (mm) of the samples.

The FM was measured as the slope of the strain versus the strain curve on the linear portion of the graph obtained

in the Bluehill 2 software (Instron Corporation, Norwood, MA, USA), using the following equation: $\text{FM} = L_1 D^3 10^{-3} / 4BH^3 D$, where: L_1 was load (N); D was distance (mm) between the supports; B was width (mm); H was height (mm); and, D was displacement (mm).

Knoop Hardness (KH)

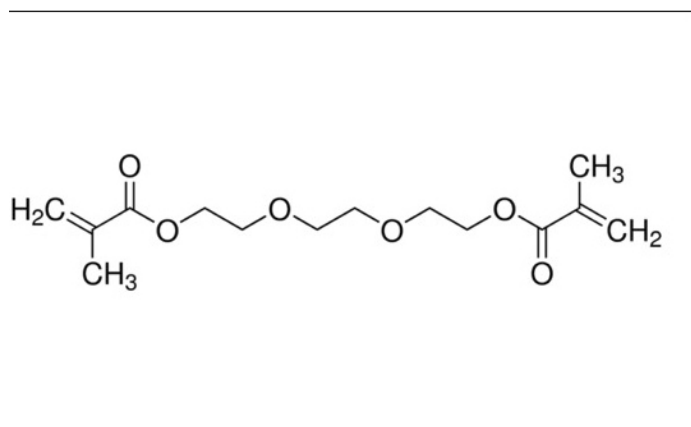
Disk-shaped samples with 2 mm in thickness and 6 mm in diameter ($n=10$) were produced from molds under the same photo-activation protocol described before. After photo-activation, the samples were removed from the molds and dry stored in light-proof containers at 37°C for 24 h . Thereafter, the top surface was wet-polished with $1,200\text{-grit SiC}$ grinding paper (Buehler, Lake Bluff, IL, USA). 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 KH value was recorded as the average of the five readings.

Solvent Degradation (SD)

After the initial KH analysis, the same disk-shaped samples ($n=10$) were used for the solvent degradation test. The samples were stored in 100% ethanol (Sigma-Aldrich Inc.) for 24 h , to calculate the KHN decrease by the softening promoted by solvent immersion using ethanol. Thus, the Knoop hardness measurements were re-taken on the same top surface after the 24 h of storage. The SD was calculated by the percentage reduction of the KH values

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Triethyleneglycol dimethacrylate
(TEGDMA)

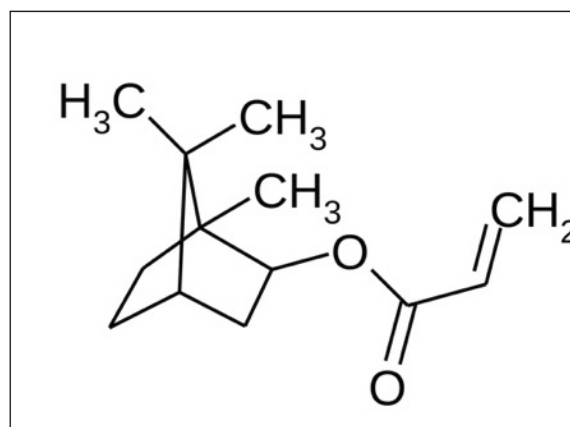


CAS Number: 109-16-0

Molecular Formula: $\text{C}_{14}\text{H}_{22}\text{O}_6$

Molecular weight: 286.32 g/mol

Isobornyl methacrylate
(IBOMA)



CAS Number: 7534-94-3

Molecular Formula: $\text{C}_{14}\text{H}_{22}\text{O}_2$

Molecular weight: 222.32 g/mol

Figure 1. Chemical structure of the diluent monomers.

after storage in relation to the KH values before storage.

Polymerization Shrinkage by Gap formation (GF) using Confocal Microscopy

The gap formation analysis was conducted using metallic molds in order to indirectly calculate polymerization shrinkage (14,15). Disc-shaped samples with 2 mm in thickness and 6.0 mm in diameter ($n=8$) were produced from metallic molds under the same photo-activation protocol described before. After photo-activation, the samples were kept in the metallic molds and dry stored in light-proof containers at 37°C for 24 h. Thereafter, the top surface of the samples was polished with 320, 400, 600 and 1200-grit SiC grinding paper (Buehler, Lake Bluff, IL, USA), and the samples stored in a 0.1% aqueous solution of rhodamine-B (Sigma-Aldrich, St. Louis, MO, USA) at room temperature for 24 h.

The samples were evaluated by Confocal Laser Microscopy (Leica TCS-SP5, Wetzlar, Hesse, Germany). A HeNe 534 laser was used for excitation, with a pass filter of 550-600nm. A 63X NA1.4 objective was used to obtain the images. The measurements (in μm) were obtained in four points located in positions corresponding to 3, 6, 9 and 12 h of a clock face. The arithmetic means was calculated considering the four points of the specimen.

Statistical Analyses

Data were checked for normality using Shapiro-Wilk's test and for variance homoscedasticity using Lavene's test. Statistical analyses were performed according to the different experimental designs with a level of significance of $\alpha=0.05$. A power analysis was previously conducted with a pilot test results to determine the sample size for each experiment in order to provide a power of at least 0.8 at a significance level of 0.5 ($\beta=0.2$). DC, FS, FM, SD and GF was analyzed using a one-way analysis of variance (ANOVA) where independent variables were set as composite formulation (containing only IBOMA as diluent monomer; combination of IBOMA and TEGDMA; or containing only TEGDMA as diluent monomer). The KH data were statistically analyzed by two-way analysis of variance (ANOVA). The factors considered were composite formulation in three

levels (60/40/0, 60/0/40 and 60/20/20) and measurement time (before and after storage in ethanol) as repeated measures. For all tests, the Tukey's test was applied for multiple comparisons between groups ($\alpha=0.05$).

Results

Table 2 shows that the diluent monomer used in the composite formulation has not influenced on the DC, FS or FM of the material. The results showed that reducing TEGDMA concentration using IBOMA either as a combining or substituent diluent monomer did not affect the DC ($p=0.805$), FS ($p=0.886$) or FM ($p=0.414$). Also, when IBOMA was used as a combining or substituent diluent monomer, lower gap formation was found in comparison to the control containing only TEGDMA as the diluent monomer ($p=0.032$). Confocal images of each formulation are illustrated in Figures 2-4.

Table 2 also shows that the diluent monomer used in the composite formulation have influenced on the KHN of the material. The combination of IBOMA and TEGDMA showed similar initial and final KHN values in comparison

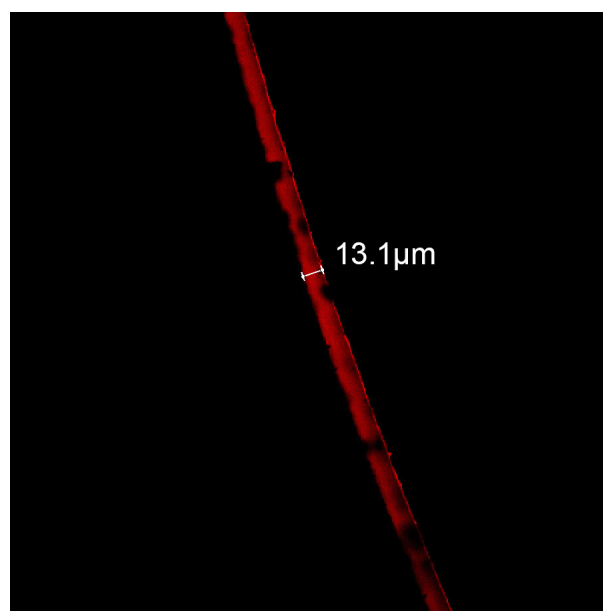


Figure 2. Confocal image showing gap formation average for the formulation containing only TEGDMA as the diluent monomer.

Table 2. DC, FS, FM, gap formation and KHN means (\pm standard deviation) for the tested resin composite formulations

TEGDMA:IBOMA ratio	DC (%)	FS (MPa)	FM (GPa)	Gap (μm)	KHN-I	KHN-F	KHN-reduction % (CS)
1:0	46.4 (3.5) A	133.0 (23.2) A	3.9 (0.5) A	11.9 (2.4) A	73.5 \pm 1.8 a,A	43.7 \pm 2.0 a,B	40.5 b
0:1	48.6 (0.5) A	136.0 (15.1) A	3.6 (0.7) A	9.3 (1.0) B	63.7 \pm 2.5 b,A	22.6 \pm 1.2 b,B	64.5 a
1:1	49.5 (1.5) A	136.5 (28.4) A	3.5 (0.6) A	9.3 (2.3) B	70.4 \pm 2.0 a,A	41.7 \pm 4.2 a,B	40.8 b

Different upper-case letters represent statistical difference in rows. Different lower-case letters indicate statistical difference in columns.

to the control group ($p=0.896$), while the total substitution of TEGDMA with IBOMA decreased the initial KHN values ($p=0.041$). Moreover, even though all composites presented reduction in KHN during the SD test, the combination of IBOMA and TEGDMA also showed similar reduction % in KHN in comparison to the control group ($p=0.001$), while the total substitution of TEGDMA with IBOMA decreased even more the KHN values ($p=0.041$).

Discussion

The first tested hypotheses that reducing TEGDMA concentration using IBOMA as a combining diluent monomer would affect the properties of resin composite formulations was rejected. As observed in the results, reducing TEGDMA concentration using IBOMA as a co-diluent monomer did not affect the DC, FS, FM, KHN or SD of the material ($p \geq 0.05$) and, actually reduced gap formation caused by polymerization shrinkage in comparison to the control containing only TEGDMA as the only diluent monomer ($p \leq 0.05$). Due to the fact that the IBOMA is a monomethacrylate monomer and do not contribute to polymer crosslinking as dimethacrylate monomers do (10,11), it can affect crosslinking but also delay the vitrification process, thus reducing shrinkage as observed.

On the other hand, the results also showed that despite totally substituting TEGDMA using IBOMA did neither affect the DC, FS or FM ($p \geq 0.05$), as well as also reduced gap formation in comparison to the control ($p \leq 0.05$), it did reduce initial KHN values as well as KHN values after SD of the formulation ($p \leq 0.05$). This means that despite providing

similar DC and some other properties in comparison to the control formulation containing only TEGDMA as the diluent monomer, totally substituting TEGDMA using IBOMA might reduce initial hardness as well as increase degradation over time. Indeed, that totally substituting the dimethacrylate monomer, TEGDMA, would culminate in no crosslinking being formed in the polymer, thus affecting KHN values and SD. Thus, the second tested hypotheses that substituting TEGDMA using IBOMA as the only diluent monomer would affect the properties of resin composite formulations was accepted. However, is worthwhile to notice that when monomethacrylate and dimethacrylate monomers were combined, crosslinking seemed not be affected and still improvements in other properties were observed.

Previous studies (12,16) evaluated monomer formulations containing isobornyl methacrylate as diluent monomer. Although these studies observed reduction in sorption and solubility, other physical-chemical properties were negatively affected, such as degree of conversion and flexural strength. It is important to point out that studies did not add filler particles in resin formulations, and the addition of filler particles does play a role in the DC as well as other physical properties of the resin material (17).

Unlike these studies, the present study evaluated the possibility to reduce TEGDMA concentration using IBOMA as a combining or substituent diluent monomer in formulations containing filler particles. The addition of particles is probably one of the most fundamental elements for the maintenance of the physical-chemical properties of resin composites (18,19). Thus, the addition

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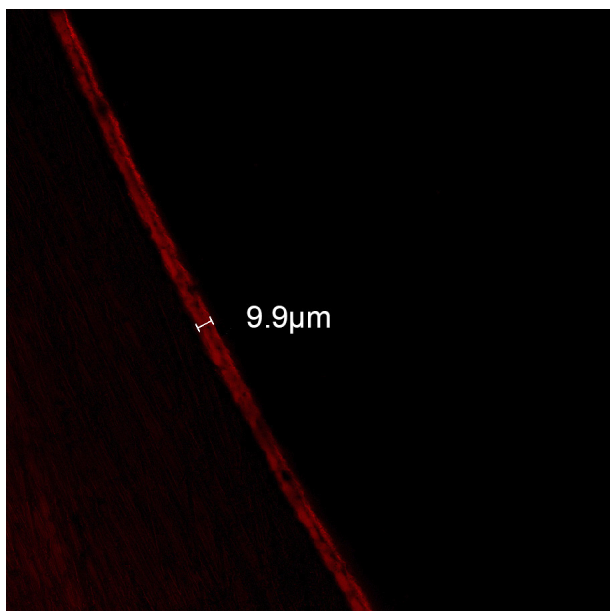


Figure 3. Confocal image showing gap formation average for the formulation containing only IBOMA as the diluent monomer.

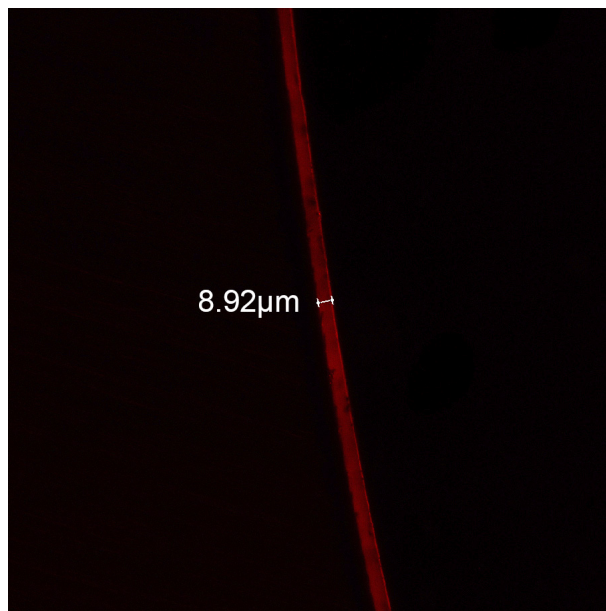


Figure 4. Confocal image showing gap formation average for the formulation containing TEGDMA and IBOMA as diluent monomers.

of this component is important to develop new composite formulations, especially when the main objectives is directly or indirectly evaluating properties such as polymerization shrinkage, gap formation and degradation potential.

As expected, the addition of IBOMA as diluent monomer led to less gap formation due to a lower polymerization shrinkage in comparison to the control group when no IBOMA was added. Although IBOMA has lower molecular weight in comparison to TEGDMA (as shown in Fig. 1), 222 g/mol and 286 g/mol, respectively, the side chain of this monomethacrylate molecule makes its mobility or diffusion more difficult during the polymerization that might consequently lead to less shrinkage and less gap formation, (12) as observed in the results.

On the other hand, it is important to point out that monomethacrylates monomers such as the IBOMA tend to form linear polymers when polymerized, (12) characterizing an amorphous structure with relatively few crosslinks and little or no crystallinity, which might directly influence on the hardness of the polymer formed (20). Therefore, it would be expected that there would be a limit in the reduction of TEGDMA and addition of IBOMA as a substituent. The TEGDMA is usually used as a cross-linking agent to co-polymerize with BisGMA in order not only to increase the DC, but also to create a chemical bond between the polymer chains formed, responsible for the physical properties of the final polymer (20). However, in general, a high crosslink density would neither be ideal because the hole-free reduction in volume among the polymer chains (20) would increase shrinkage and gap formation.

Thus, it was possible to verify that it is possible to reduce TEGDMA concentration using IBOMA as a combining diluent monomer in composite formulations. However, it was not possible to totally substitute TEGDMA using IBOMA. There is limit in the reduction of TEGDMA and addition of IBOMA as a substituent in order not to influence on the physical properties of the material, but a 1:1 ratio combination does seem suitable to be used in composite formulations without affecting physical or chemical properties and decreasing potential gap formation, which would predict better longevity over time. Thus, within the limitations of this study, it was possible to conclude that the combination of IBOMA and TEGDMA seem to be suitable, allowing the reduction of TEGDMA concentration without affecting the properties of resin composite formulations, and actually reducing polymerization shrinkage as well as solvent degradation, thus potentially reducing cytotoxic potential of resin composite formulations containing IBOMA as a combining diluent monomer.

Resumo

O objetivo nesse estudo foi utilizar o isobornil metacrilato (IBOMA) como

monômero combinante ou substituinte na matriz resinosa de resinas compostas odontológicas. Para tanto, a matriz resinosa foi formulada com 60 % em peso de BisGMA e 40 % em peso de monômero diluente. O TEGDMA foi utilizado como único monômero diluente para o grupo controle com 40 % em peso, enquanto a substituição total de TEGDMA foi feita com 40 % em peso de IBOMA. Na combinação de IBOMA e TEGDMA foram utilizados 20 % em peso de cada monômero. Na matriz resinosa, 65 % em peso de partículas de carga foi adicionado. O grau de conversão (GC) utilizando FT-IR, a resistência flexural (RF), o módulo flexural (MF), a contração de polimerização por formação de gap (FG) utilizando Microscopia Confocal à Laser, a Dureza Knoop (DK) e a degradação em solvente (DS) foram avaliados. Todos os dados foram analisados utilizando ANOVA e teste de Tukey ($\alpha=0.05$; $\beta=0.2$). Os resultados demonstraram que reduzindo ou substituindo o TEGDMA utilizando o IBOMA não afetou o GC ($p=0.085$), a RF ($p=0.886$) ou o MF ($p=0.414$). Ainda, quando o IBOMA foi utilizado como um monômero combinante ou substituinte, menor FG foi observada em comparação ao controle contendo apenas TEGDMA como monômero diluente ($p=0.032$). Contudo, apesar de todos os compósitos terem apresentados redução na DK durante o teste de DS, a combinação de IBOMA e TEGDMA demonstrou uma redução na DK similar ao grupo ($p=0.001$), enquanto a substituição total de TEGDMA com IBOMA reduziu a DK após DS ($p=0.041$). Dessa forma, a combinação do IBOMA e TEGDMA parece reduzir DS e a FG sem afetar as propriedades de resinas compostas.

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