



**Tatiany Gabrielle Freire Araújo**

“Influência de solventes nas propriedades físico-químicas de infiltrantes resinosos experimentais”

“Influence of solvents on the physicochemical properties of experimental resin infiltrants”

**Piracicaba  
2015**





**Universidade Estadual de Campinas  
Faculdade de Odontologia de Piracicaba**

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infiltrantes resinosos experimentais”**

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Tese apresentada à Faculdade de Odontologia de Piracicaba da Universidade Estadual de Campinas como parte dos requisitos para obtenção do título de Doutora em Materiais Dentários.

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Orientador: Prof. Dr. Américo Bortolazzo Correr

Este exemplar corresponde à versão final da Tese defendida pela aluna Tatiany Gabrielle Freire Araújo e orientada pelo Prof. Dr. Américo Bortolazzo Correr

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## RESUMO

O objetivo neste estudo foi avaliar o efeito da adição dos solventes dimetil sulfóxico (DMSO) e tetrahidrofurano (THF) em propriedades físico-químicas de infiltrantes experimentais. Foram preparadas duas blendas resinosas, uma composta por 75%p de TEGDMA (T) e 25%p UDMA (U) e outra por 75%p de T e 25%p de BISEMA (B). Em um grupo de cada mistura não foi adicionado solvente. Os demais grupos foram formulados pela adição dos solventes DMSO ou o THF nas concentrações de 0,5% e 5%. No total foram obtidos 10 infiltrantes experimentais. Icon® foi utilizado como controle. Este estudo foi separado em 2 capítulos. No capítulo 1 o objetivo foi avaliar a influência dos solventes DMSO e THF no ângulo de contato, dureza Knoop em lesões artificiais de cárie infiltradas e a capacidade de penetração dos infiltrantes experimentais em microscópio confocal de varredura a laser. Os dados do ângulo de contato foram submetidos à análise de variância um fator e teste de Tukey, e os de dureza Knoop avaliados por análise de variância dois fatores e teste de Tukey ( $\alpha=0,05$ ). Os resultados mostraram que o Icon apresentou o menor ângulo de contato. Dentre os infiltrantes experimentais, o menor ângulo de contato foi apresentado pela mistura T + U + 5% DMSO, significativamente menor que as misturas T + U, T + U + 0,5% DMSO, T + U + 5% THF, T + B + 0,5% DMSO e T + B + 0,5% THF. Os resultados de microscopia confocal de varredura a laser mostraram que os infiltrantes apresentaram boa penetração nas lesões artificiais de cárie exceto os grupos T + B, T + B + 0,5% DMSO e T + B + 0,5% THF. As lesões infiltradas por Icon apresentaram dureza significativamente maior que dos outros materiais, exceto T + U + 0,5% DMSO (252,4) e T + U + 5% THF (239,1). Concluiu-se que o Icon apresentou os melhores resultados dentre os materiais avaliados, com menor ângulo de contato, maior dureza e boa penetração. Dentre os materiais experimentais, a incorporação de 5% de DMSO na mistura de U reduziu o ângulo de contato e apresentou dureza semelhante à infiltrada por Icon. No capítulo 2 foi verificada a influência dos solventes DMSO e THF nos infiltrantes experimentais no grau de conversão (GC) (n = 3), resistência coesiva à tração (RT) (n = 10), resistência de união a microtração (RU) (n = 10), resistência a flexão (RF) (n = 10) e módulo de elasticidade (ME) (n = 10). Os dados foram submetidos à ANOVA um fator e



teste de Tukey ( $\alpha = 0,05$ ). Foi verificado que o GC dos infiltrantes com U foi significativamente maior que dos demais infiltrantes. Os infiltrantes experimentais sem solvente apresentaram RF significativamente maior que Icon e que os infiltrantes com solvente. Os infiltrantes com 5% de DMSO apresentaram RU significativamente menor que os demais; Icon apresentou RU significativamente maior que os demais infiltrantes. A RT de Icon foi significativamente maior que dos infiltrantes experimentais. Conclui-se que os solventes DMSO e THF não melhoraram a resistência de união e prejudicaram as propriedades mecânicas dos infiltrantes experimentais. Dentre os solventes, o THF na concentração de 0,5% apresentou menos efeitos deletérios nas propriedades mecânicas dos infiltrantes experimentais. Como conclusão geral, pode ser verificado que o infiltrante Icon apresentou os melhores resultados. Dentre os infiltrantes experimentais, o melhor desempenho foi obtido com a mistura T e U. A adição 0,5% do solvente DMSO na mistura T e U não reduziu o ângulo de contato e reduziu as propriedades mecânicas dos infiltrantes, mas produziu resistência de união e dureza da lesão infiltrada semelhante ao infiltrante Icon.

**Palavras-chave: solventes, metilmetacrilato, infiltração.**

## ABSTRACT

The aim of this study was to evaluate the effect of alternative solvents addition dimethyl sulfoxide (DMSO) and tetrahydrofuran (THF) in the chemical-physical properties of experimental infiltrants. Were prepared 2 blends: (1) 75wt% TEGDMA (T) + 25 wt% UDMA (U), (2) 75wt% T + 25wt% BISEMA (B). From each blend were added the solvents DMSO or THF in concentrations of 0.5wt% and 5wt%, totaling 10 experimental groups. Icon® was used as control. This study was divided in 2 chapters. In chapter 1 the aim was to evaluate the influence of alternative solvents (DMSO and THF) in the contact angle, Knoop hardness of artificial caries lesions infiltrated, and the capability of penetration of the experimental infiltrants. Data were analyzed by ANOVA one-way (contact angle) and two-way (Knoop hardness) and Tukey's test ( $\alpha=0.05$ ). The results showed that Icon presented the lowest contact angle. Analyzing the experimental infiltrants, T + U + 5% DMSO showed the lowest contact angle. The confocal microscopy analysis showed that the infiltrants presented satisfactory penetration into the caries-like lesions except the groups T + B, T + B + 0.5% DMSO and T + B + 0.5% THF. Lesions infiltrated with Icon exhibited hardness values significantly higher than all the experimental groups, except to T + U + 0,5% DMSO (252.4) e T + U + 5% THF (239.1). It is possible to conclude that Icon showed the best results for the tested properties. In chapter 2 was analyzed the influence of the addition of DMSO and THF solvents in experimental infiltrants in relation to degree of conversion (n=3), ultimate tensile strength (n=10), microtensile bond strength (n=10), flexural strength (n=10) and elastic modulus (n=10). Data were submitted to one-way ANOVA and Tukey's test ( $\alpha=0.05$ ). Infiltrants containing U obtained the highest degree of conversion values. The solvents-free experimental infiltrants showed the highest values of flexural strength. In relation to microtensile bond strength results, the groups with 5% DMSO presented the lowest results and Icon the highest ones. Icon group obtained the highest results of UTS. It is possible to conclude that the addition of solvents DMSO and THF did not improve the bond strength and affected negatively mechanical properties of the experimental infiltrants. THF solvent 0,5% showed less deleterious effects in mechanical properties of experimental infiltrants. Overall, Icon

obtained the best results. Among the experimental infiltrants, the best performance was obtained by the association of T and U. The addition of 0.5% of DMSO in the blend T + U did not reduce the contact angle and affected negatively the mechanical properties, except to microtensile bond strength and hardness of the caries lesions infiltrated.

Key- words: solvent, monomer, infiltrant.

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## ***Epígrafe***

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*“Os que se tornam sábios são felizes, e a sabedoria lhes dará vida”  
Provérbios 3:18*



## INTRODUÇÃO

As lesões cariosas incipientes do esmalte dental, geralmente denominadas lesões de mancha branca, apresentam-se como área de desmineralização da camada sub-superficial do esmalte (Bergman e Lind, 1966). Quando a mancha branca é identificada clinicamente, os cristais de hidroxiapatita na subsuperfície apresentam redução em volume e os espaços ou poros intercristalinos aumentam (Bergman e Lind, 1966). Essas áreas cariadas apresentam resistência mecânica alterada devido à desmineralização. O tratamento com fluoretos pode não devolver as características do esmalte sadio, pois a remineralização não é homogênea em toda a extensão da lesão (Mejare *et al.* 1998).

A penetração e a paralisação de lesões artificiais de cárie por adesivos e selantes têm sido avaliadas em vários estudos laboratoriais (Rodda, 1983; Donly e Ruiz, 1992; Robinson *et al.*, 2001; Gray e Shellis, 2002; Schmidlin *et al.*, 2004; Meyer-Lueckel *et al.*, 2006; Paris *et al.*, 2006). Entretanto, selantes e adesivos não são materiais ideais para maior penetrabilidade, mostrando apenas penetração superficial nas lesões de esmalte natural (Paris *et al.*, 2007b).

Em adesivos, a água, o etanol e a acetona são os solventes mais comumente utilizados (Van Landuyt *et al.*, 2007). Em sistemas adesivos de técnica úmida, o solvente melhora a difusão dos monômeros dentro da matriz colágena exposta da dentina (Pashley *et al.*, 1995). A fim de melhorar as propriedades dos materiais, um estudo prévio (Paris *et al.* 2007a) foi realizado variando a combinação de monômeros e foram encontrados maiores valores de coeficiente de penetração (CP) a partir de combinações com maiores teores de HEMA, TEGDMA e etanol. Em contraste, a incorporação de BisGMA e UDMA aumentou a viscosidade e diminuiu o CP do material. Paris *et al.* (2007a) encontraram que misturas com a adição de etanol apresentaram o maior CP (470 cm/s) devido a menor viscosidade e ângulos de contato. Entretanto, apesar do favorável maior CP, composições contendo HEMA e etanol mostraram incompleta polimerização (Paris *et al.* 2007a; Araújo *et al.* 2013), além de resultar na diminuição das propriedades dos materiais experimentais (Araújo *et al.*, 2013). Conseqüentemente, a adição de HEMA e etanol em infiltrantes resinosos pode causar efeitos deletérios, devendo ser cautelosamente realizada. Paris e

Meyer Lueckel (2010c) concluíram em estudo *in vitro* que resinas de baixa viscosidade (infiltrantes) à base de TEGDMA apresentando coeficiente de penetração relativamente alto (>200 cm/s) foram capazes de inibir quase que completamente a progressão de lesões de cárie naturais em esmalte em meio desmineralizante.

Foi introduzido no mercado odontológico o material infiltrante Icon<sup>®</sup> (DMG; Hamburg, Germany) com propósito de infiltrar as lesões iniciais de cárie. Esse material, com base no TEGDMA, tem sido estudado e apresentado êxito para paralisação da progressão de lesões de cárie em esmalte. Paris *et al.* (2010b) após 18 meses de avaliação obtiveram resultados satisfatórios, demonstrando que a progressão das lesões de cárie interproximal *in vivo* foi interrompida. Entretanto, devido às características de sorção de água apresentada pelo TEGDMA, outras combinações monoméricas, incorporadas com diferentes solventes devem ser estudadas, para avaliar a longevidade dos materiais.

O dimetil sulfóxido (DMSO) é um solvente amplamente utilizado em muitos tipos de reações químicas que requerem maior polaridade, assim como em métodos analíticos e aplicações práticas de bioquímica e farmacologia (Vaisman e Berkowitz, 1992; Chalaris *et al.*, 2008). É um solvente com tendência em formar associações intermoleculares com si próprio e com outras moléculas, tais como a água (Shikata e Sugimoto, 2011). Bioquimicamente, é uma molécula anfipática, ou seja, apresenta a característica de possuir uma região hidrófila (solúvel em meio aquoso) e uma região hidrófoba (insolúvel em água; porém, solúvel em lipídios e solventes orgânicos), sendo um dos solventes mais comumente utilizados para substâncias insolúveis em água (Pal *et al.* 2011).

O tetrahidrofurano (THF) é um solvente orgânico heterocíclico aprótico polar, sendo que estes solventes são bons para dissolver ampla gama de massas polares e apolares (íons) (Graham e Fryhle, 2001). Estas características podem justificar o uso de THF em adesivos dentários que combinam moléculas hidrófobas e hidrófilas (Van Landuyt *et al.*, 2007). Além disso, a característica volátil do THF implica que este solvente pode ser facilmente removido depois da aplicação (Reis *et al.*, 2008). Fontes *et al.* (2009) concluíram que THF é um solvente promissor para o uso em sistemas adesivos devido à estabilidade da união ao longo do tempo quando este solvente é adicionado aos sistemas

adesivos. Em relação a citotoxicidade, THF mostrou resultados promissores *in vitro*, além de apresentar alta estabilidade na interface adesiva após um ano de armazenamento em água (Fontes, *et al.*, 2013). De modo geral, esses solventes podem auxiliar na capacidade de penetração dos materiais resinosos.

Novos materiais devem ser desenvolvidos para obter penetração eficiente e oferecer estrutura ácido-resistente pós-polimerização, reforçar a estrutura da lesão cariiosa, impedir a progressão da cárie, aumentar e melhorar a longevidade dos materiais. Assim, seria interessante incorporar aos materiais infiltrantes experimentais monômeros associados a solventes que não interfiram na polimerização e estabilidade do polímero ao longo do tempo, além de diminuir uma etapa clínica presente no material comercialmente disponível. Portanto, o objetivo neste estudo foi avaliar as propriedades físico-químicas de infiltrantes resinosos contendo diferentes monômeros e solventes, bem como a união destes materiais ao esmalte e sua capacidade de penetração em lesões artificiais de cárie em esmalte.

### CAPÍTULO 1

**Influence of monomers and solvents DMSO and THF on the contact angle, penetrability and Knoop hardness of experimental resin infiltrants into caries-like lesions**

#### ABSTRACT

**Objective:** To evaluate the influence of monomers urethane dimethacrylate (UDMA) and ethoxylated bisphenol A glycidyl dimethacrylate (BisEMA) and solvents tetrahydrofuran (THF) and dimethyl sulfoxide (DMSO) on contact angle, penetrability and Knoop hardness of resin infiltrants into caries-like lesions. **Methods:** Eleven groups were evaluated: Icon; 75% TEGDMA (T) + 25%UDMA (U); 75%T + 25%U + 0,5%DMSO; 75% T + 25% U + 5% DMSO; 75% T + 25% U + 0,5% THF; 75% T + 25% U + 5% THF; 75% T + 25% BisEMA (B) ; 75% T + 25% B + 0,5% DMSO; 75% T + 25% B + 5% DMSO; 75% T + 25% B + 0,5% THF; 75% T + 25% B + 5% THF. The contact angle measurement was performed in a viscosimeter. The resin infiltrants penetrability and Knoop hardness was evaluated in caries-like lesions produced on enamel of bovine teeth. Contact angle data were submitted to one-way ANOVA and Tukey's test. Knoop hardness was evaluated by two-way ANOVA and Tukey's test. It was used 5% significance level. Penetrability was evaluated in Confocal Laser Scanning Microscopy by qualitative analysis. **Results:** Icon showed contact angle significantly lower than the other groups. T+U+5DMSO showed contact angle significantly lower than T+U; T+U+0.5DMSO; T+U+5THF; T+B+0.5DMSO; and T+B+0.5THF. Confocal Laser Scanning Microscopy showed good penetration for all resin infiltrant. Caries-like lesions infiltrated by Icon showed Knoop hardness significantly higher than other groups, except for the T+U+0.5DMSO and T+U+5THF groups. The Knoop hardness significantly increased in deeper sites. It can be concluded that Icon showed best performance between resin infiltrants (lower contact angle, higher Knoop hardness and good penetration).

**Key-words:** contact angle, Knoop hardness, confocal microscopy, solvent, infiltrant.

### INTRODUCTION

The resin infiltration technique is a promising approach to reinforce demineralized enamel. With the use of adequate materials with a sufficient resistance to mechanical and chemical degradation, and with a careful application technique, lesion arrest seems achievable on a supervised basis, thus closing the gap between oral hygiene and minimally invasive dentistry, and providing a wait-and-see position to both the clinician and the patient<sup>1</sup>. The caries infiltration technique aims to occlude the pores within the lesion body with resin, impeding the continuous diffusion of acids and dissolved minerals through the lesion, and thus hampering caries progression<sup>2</sup>. Initial lesions in smooth surfaces infiltrated with low-viscosity resins became more resistant to further demineralization, and the ability to inhibit lesion progression was strongly correlated with the penetration depth (PD) of the resin<sup>2</sup>.

Materials based on methacrylate have been developed and studied to penetrate the lesion body and reinforce the weakened structure of enamel caries lesions<sup>3</sup>. Thus, some characteristics such as low viscosity, low contact angle and low surface tension are essential in order to allow a deep penetration of the material into the incipient caries lesions. Paris *et al.* (2007)<sup>4</sup> using resin blends with different formulations from those used as pit and fissure sealants, or adhesive resin, proposed optimized materials to infiltrate enamel carious lesions, the so called "infiltrants". The penetration of infiltrants into the pores of the lesion body is mainly driven by capillary forces and depends on the penetration time, the capillary radius and the penetration coefficient (PC) of the liquid<sup>5</sup>.

The PC combines the liquid properties viscosity, surface tension and contact angle to the solid<sup>6</sup>. Materials with different PCs and different concentrations of ethanol were tested in natural caries lesions in permanent teeth and deepest and most homogeneous infiltration was obtained with a solvent-free resin<sup>7</sup>. Also was found that the addition of solvent ethanol or HEMA into TEGDMA blends does not improve the penetration depth and homogeneity of the infiltrants<sup>8</sup>. Characteristic parameters like resin penetration depth



or polymerization shrinkage as well as hardness of infiltrated lesions might be influenced by monomer and solvent composition of infiltrants<sup>9</sup>.

It was introduced in the dental market the infiltrating material Icon<sup>®</sup> (DMG, Hamburg, Germany) for the purpose of infiltrating the incipient carious lesions. This material based on TEGDMA, has been studied and shown success. Paris *et al.* (2010)<sup>10</sup>, after evaluation over 18 months showed satisfactory results, demonstrating that progression of lesions *in vivo* interproximal caries was interrupted. The current results corroborate that caries infiltration is an efficacious method to hamper progression of noncavitated proximal lesions extending radiographically into the inner half of enamel up to the outer third of dentin after a period of 3 years.<sup>11</sup> The addition of alternative solvents can improve the stability of infiltrating as well as adhesives<sup>12</sup> and reduced the solvent clinical application time recommended by the manufacturer of infiltrant Icon. The solvent dimethyl sulfoxide (DMSO) is used in various biochemical and pharmacological practice due to its high polarity<sup>13,14</sup>. Likewise, tetrahydrofuran (THF) was reported promising as an alternative solvent and due to their advantageous volatile properties<sup>15</sup>. So these are promising solvents can besides increasing the viscosity of infiltrants, promote better infiltration in the body of lesion.

The aim of the present study was to evaluate the influence of solvents THF or DMSO used in experiments infiltrants materials on properties (contact angle and Knoop hardness) and to evaluate the penetration depth of the materials into enamel caries lesions. The null hypotheses tested were:

- 1- There would no differences on the Knoop hardness of lesions caries infiltrated by different resin materials.
- 2- There would no differences on the contact angle of the different resin materials.
- 3- There would no differences on the infiltration in lesions caries by the infiltrants.

### MATERIAL AND METHODS

#### *1. Infiltrant preparation*

To prepare the infiltrants, the following monomers were used: triethyleneglycol dimethacrylate (TEGDMA) (Sigma-Aldrich Inc., Batch #01612M), ethoxylated bisphenol A glycidyl dimethacrylate (BisEMA) (Sigma-Aldrich Inc., Batch #03514HF), urethane dimethacrylate (UDMA) (Sigma-Aldrich Inc., Batch #09405B), Dimethyl Sulfoxide (DMSO), tetrahydrofuran(THF) (Sigma-Aldrich Inc., Batch #51496AM) in different ratios, as described in Table 1.

For each experimental group, monomers resin blend were mixed up in brown glass jars. For all blends, the photoinitiator system selected was camphorquinone (CQ) (Sigma-Aldrich, Batch #532604) as photoinitiator and dimethyl aminoethyl methacrylate (DMAEMA) (Sigma-Aldrich Inc., Batch #BCBF8391V) as co-initiator (proportion 1:2 by weight). The light-curing initiator system was thoroughly dissolved in the monomer matrix with the concentration of 1.5 wt% (0.5% CQ/ 1% DMAEMA), Also, the inhibitor butylated hydroxytoluene (BHT) (Sigma-Aldrich Inc., Batch #04416KD) was added to the resin blends with a concentration of 0.1 wt% in order to avoid the spontaneous polymerization of the monomers. In order to avoid premature polymerization, the resin blend groups were stored at 4°C until use.

Table 1-Infiltrant blends composition.

<b>Infiltrant</b>	<b>Composition (wt,%)</b>
Icon <sup>®</sup>	Methacrylate resin
T + U	75% TEGDMA +25%UDMA
T + U + 0.5% DMSO	T+U+0.5%DMSO
T + U + 5% DMSO	T+U +5%DMSO
T + U + 0.5% THF	T+U +0.5%THF
T + U + 5% THF	T+U +5%THF
T + B	75% TEGDMA+ 25% BisEMA
T + B + 0.5% DMSO	T+B+0.5%DMSO
T + B + 5% DMSO	T+B+5%DMSO
T + B + 0.5% THF	T+B +0.5%THF
T + B + 5% THF	T+B +5%THF

T- triethyleneglycol dimethacrylate;B- ethoxylated bisphenol A glycidyl dimethacrylate; U- urethane dimethacrylate; DMSO- Dimethyl Sulfoxide; THF-tetrahydrofuran.

## **2. Contact angle**

The contact angle measurements were performed on glass slides polished. To measure the contact angles of resin infiltrants a viscometer (GBX Instrumentation Scientifique -France No, 04230711- Contact Angle Meter - Digitrop) with an attached camera was used. Droplets of liquid materials (about 1µl) were placed on a glass slide via a micro syringe. After 10 s, an image was recorded and analyzed using a software drop shape analysis. For each group (n = 5), the average of three measurements was calculated. Each measurement was performed on a new glass slide. Data was statistically analyzed by one-way ANOVA and Tukey’s multiple comparison test ( $\alpha = 0.05$ ).

## **3. Knoop hardness (KHN) and Confocal Laser Scanning Microscopy (CLSM)**

### **3.1. Specimen preparation**

One hundred and sixty five bovine incisors without cracks or caries were collected. The teeth were cleaned and stored in 0.1% thymol solution up to 1 month after

extraction. The roots were sectioned 1 mm below the cement enamel junction using a diamond disc (Isomet, Buehler Ltd., Lake Bluff, IL) and discarded. The teeth were cut using a diamond disc (Isomet Buehler Ltd., Lake Bluff, IL) to obtain blocks of enamel (5 x 5 mm). Enamel surfaces of fifty five teeth (n = 5) were ground flat with water-cooled mechanical grinding machine using silicon carbide discs 340- and 600-grit Al<sub>2</sub>O<sub>3</sub> abrasive paper (Aropol E, Arotec S.A. Ind.&Com., São Paulo, Brazil) for analyses in CLSM. The other one hundred and ten teeth were used to assess the Knoop Hardness (n = 10).

### ***3.2. Artificial enamel caries-like lesion formation***

The subsurface enamel caries-like lesions (ECLL) were produced in the sound enamel surface. Each enamel block was covered with double coats of acid-resistant nail varnish (Colorama<sup>®</sup> - São Paulo, Brazil) except to the polished enamel area (5 x 5 mm). The ECLL was produced by immersion of each enamel surface into 50 mL of a demineralizing solution containing 0,05 M acetate buffer 50% hydroxyapatite saturated from enamel powder, pH 5.0, for 16 h at 37 °C<sup>16</sup>. To prepare the solution, enamel powder (particles of 74-105 µm) was agitated into 0.05 sodium acetate buffer, pH 5.0, for 96 h at 37°C (0.50 g/L)<sup>17,18</sup>. The solution was used in a ratio of 2.0 mL/mm<sup>2</sup> of exposed enamel area. Immersion period of 16 h was determined in a previous study, by analyzing thin enamel slices with polarized light microscopy. It was clearly observed the presence of subsurface ECLL. Calcium concentration into the solution was 66.3 µg/mL, which was determined by atomic absorption spectrometry with flame spectrophotometer model 506 (Perkin Elmer); phosphorus concentration was about 32 µg/mL, which was determined by colorimetric method of Fiske and Subbarow (1925) with spectrophotometer model 800M (Analyzer) adjusted at 660 nm.

### ***4. Lesion infiltration and preparation for CLSM***

The enamel blocks with ECLL were randomly distributed into eleven groups (Table 1) (n = 5). The enamel was etched with 37% phosphoric acid gel (Magic Acid, Vigodent, Rio de Janeiro, Brazil) for 60 s<sup>19</sup>, washed with water spray, and dried for 15 s. Infiltrants were impregnated with 0,1% rhodamine B isothiocyanate (RITC) (Sigma-

Aldrich), applied onto the caries-like lesion using a microbrush, and left to penetrate for 60 s<sup>20</sup>. Block surface was air-dried for 15 s in order to evaporate the solvent. All groups were light cured for 60 s with Ultralume 5 (Ultradent, South Jordan, Utah, USA) with 1000 mW/cm<sup>2</sup> irradiance. Tooth sections with 0.5 mm thickness were obtained perpendicularly to the lesion surface, impregnated with the materials using a diamond saw (Isomet 1000, Buhler, Lake Bluff, IL, USA), and polished in a SiC papers series (#600, # 1200, # 2000). To visualize the porous structure (not infiltrated lesion parts), specimens were immersed in a 50% ethanolic solution of 100 mM sodium fluorescein (NaFl) (Sigma Aldrich) for 3 min and washed in deionized water for 10 seconds<sup>8</sup> (Figure 1).

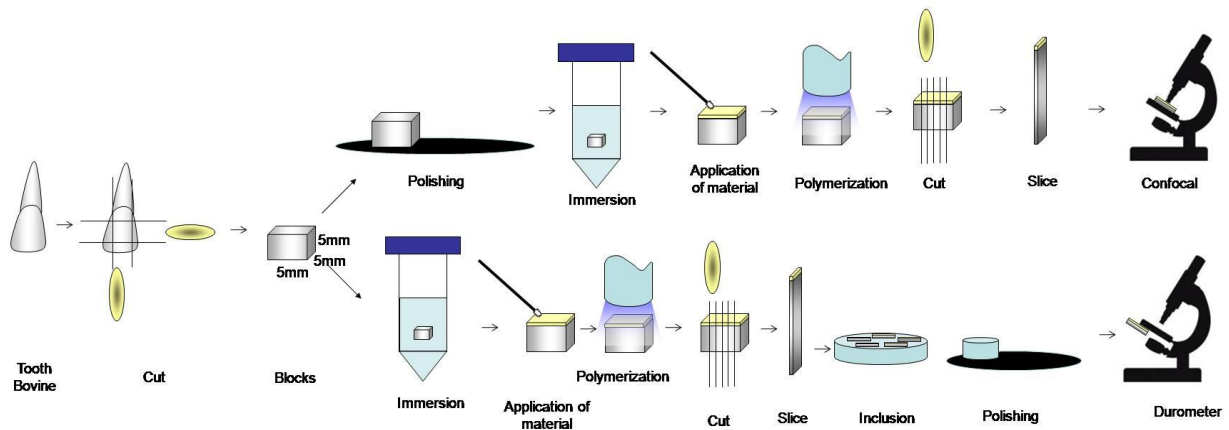


Figure 1. Representative scheme showing the specimen preparation for CLSM and KHN.

### 4.1. CLSM evaluation

Specimens were observed with a confocal laser scanning microscope (Leica, TCS NT; Leica, Heidelberg, Germany) using a 10 x objective in dual fluorescence mode as described previously<sup>21</sup>. The excitation light had two wavelength maxima, at 488 and 568 nm. The emitted light was split by a 580-nm reflection short-pass filter and was passed through 525/50-nm band-pass filter for rodhamine B and a 590-nm long-pass filter for rodhamine B detection.

Images with a lateral dimension of 1.0 X 1.0 mm<sup>2</sup> and a resolution of 1024 X 1024 pixels were recorded and analyzed by Leica SP2 CLSM software (Zeiss, Oberkochen,

Germany). The analysis was blind with regard to the group allocation of the teeth. A qualitative analysis was employed to evaluate the penetration of materials into the lesion body.

### **5. Lesion infiltration and preparation for KHN**

The enamel blocks with ECLL were randomly distributed into eleven groups (Table1) (n=10) according to the composition of low viscosity resin materials. The previously determined area (5x5mm) on enamel surface blocks was etched with 37% phosphoric acid gel (Vigodent, Rio de Janeiro, RJ, Brazil) for 60 seconds<sup>19</sup>, rinsed for 10 seconds and air-dried for 15s. The experimental infiltrants were applied using micro-brush for 60s<sup>20</sup> in order to improve the penetration into the etched enamel. Block surface was air-dried for 15 s in order to evaporate the solvent. Infiltrants were then light cured for 60s with Ultralume 5 (Ultradent, South Jordan, Utah, USA) with 1000mW/cm<sup>2</sup>. Blocks of experimental infiltrants with 5 mm height (increments of 2 mm-thickness) were done using a silicon mold and individually light cured for 60s. Then, the enamel blocks were stored in 100% humidity for 24 h at 37°C. Afterwards, each enamel block was longitudinally cut into slices of 2 mm thickness using water-cooled diamond blade (Isomet 1000 - Buehler Ltd., Lake Bluff, IL, USA). The slices of all groups were embedded in acrylic resin in a polymethyl methacrylate resin (PVC) matrix and polished with water-cooled silicon carbide discs (600, 1200, 2000, grade papers) (Figure 1).

#### **5.1. KHN evaluation**

The longitudinal KHN measurement was performed through three sequences of four indentations at distances of 10µm, 30µm, 50 µm and 100 µm from the surface under a load of 490 N (50g) for 10 seconds using the microhardness tester (HMV-2, Shimadzu, Tokyo, Japan). The measures of hardness were calculated in each distance (kgf/mm<sup>2</sup>). The results were statistically analyzed using two-way ANOVA and Tukey's test ( $\alpha = 0.05$ ).

**RESULTS**

The two-way ANOVA for hardness showed significant differences for infiltrants ( $p < 0.001$ ) and depth ( $p < 0.001$ ) factors, but not for the interaction between factors ( $p = 0.56$ ). The hardness outcomes (means and standard deviations) of different resin infiltrants are depicted in Table 2.

Statistically significant differences ( $p < 0.001$ ) were found between the different monomers and solvents tested in this study. The resin infiltrants T+B+5THF and T+B obtained the lowest KHN (respectively 148.3 and 160.8) whereas T+B+5DMSO (175.9); T+U (189.2); T+U+0.5%THF (192.6); T+B+0.5DMSO (205.6); T+U+5DMSO (222.6); and T+B+0.5%THF (226.6) presented intermediary outcomes. The resin infiltrants Icon (266.4); T+U+0.5DMSO (252.4); T+U+5THF (239.1) showed the highest KHN. The UDMA promoted significantly higher Knoop hardness compared to BisEMA, except for group 0.5%THF. The mean values varied from the lowest (148.9) Knoop hardness for T+B+5THF to the highest for Icon<sup>®</sup> (266.4) (Table 2).

Table 2- Mean (standard deviation) values of Knoop hardness of infiltrated lesions.

Groups	Hardness (KHN)
Icon	266.4 (50.7) a
T+U	189.2 (47.1) cd
T+U+0.5DMSO	252.4 (67.6) ab
T+U+5DMSO	222.6 (57.7) bc
T+U+0.5THF	192.6 (48.1) cd
T+U+5THF	239.1 (43.9) ab
T+B	160.8 (58.3) ef
T+B+0.5DMSO	205.6 (39.7) c
T+B+5DMSO	175.9 (37.4) de
T+B+0.5THF	226.6 (44.4) bc
T+B+5THF	148.3 (31.9) f

Different letters indicate statistically significant difference ( $p < 0.05$ ).

## Capítulo 1

The Knoop hardness increased significantly from the depth of 10 micrometers up to 100 micrometers (Table 3).

Table 3- Mean (standard deviation) values of Knoop hardness in different depth into the caries-like lesions.

Depth	Knoop hardness		
10 micrometer	172.5	55.1	D
30 micrometer	190.4	49.5	C
50 micrometer	218.0	52.5	B
100 micrometer	247.9	55.9	A

Different letters indicate statistically significant differences ( $p < 0.001$ ).

The contact angle outcomes are presented in Table 4. ANOVA showed significant differences between groups ( $p < 0.001$ ).

The contact angle of Icon<sup>®</sup> (11.4) was significantly lower than all experimental infiltrants. The T+U+5DMSO experimental resin infiltrant showed contact angle significantly lower than T+U; T+U+0.5DMSO; T+U +5THF; T+B+0.5DMSO; and T+B+0.5THF. There were no significant differences among other six groups (Table 4).

Table 4- Mean (standard deviation) values of contact angle of different infiltrants.

Groups	Contact angle (°)
Icon	11.4 (2.4) c
T + U	29.8 (6.3) a
T + U + 0.5DMSO	29.5 (5.5) a
T + U + 5DMSO	16.7 (3.3) b
T + U + 0.5THF	23.0 (1.3) ab
T + U + 5THF	31.8 (3.7) a
T + B	26.4 (3.5) ab
T + B + 0.5DMSO	32.3 (5.7) a
T + B + 5DMSO	24.5 (6.8) ab
T + B + 0.5THF	29.8 (3.6) a
T + B + 5 THF	25.4 (0.5) ab

Different letters indicate statistically significant differences ( $p < 0.05$ ).



According to Confocal Microscopy evaluation, a qualitative analysis was employed to evaluate the penetrated materials into the lesion body. Confocal micrographs are presented in Figures 3, 4 and 5.

In figure 2 is shown the penetrability of commercial infiltrant Icon<sup>®</sup>. Note on the figure 2a the demineralized area. In the image 2b is observe the infiltrant impregnated with rodhamine b into the pores of enamel caries. Sequentially in image 2c demonstrated the overlapping of the demineralized areas and red areas, indicating the infiltrant impregnated with rodhamine b into the lesion body. In this group can be observed that the full extent of the lesion was penetrated by commercial infiltrant Icon<sup>®</sup>.

The penetration of experimental infiltrants with base blend TEGDMA + UDMA into the demineralized areas is showed in figure 3. Note on the images 3a for all groups the demineralized area by the artificial caries lesions. In the figure 3b can be observed the infiltrants impregnated with rodhamine b. All groups with base blend TEGMA + UDMA showed good penetration into the lesion body. Note the images 3c in the overlapping of the demineralized areas and red areas, the body of lesions totally infiltrated by the experimental material.

In the figure 4 it is showed the penetration of mixtures TEGDMA and BisEMA. The images 4a presents the demineralized area for all groups. Note on the images 4b for all groups the experimental infiltrants impregnated with rodhamine b. The overlapping of the demineralized areas and red areas is observed in images 4c for all groups. In groups T + B; T + B + 0.5% DMSO and T + B + 0.5% THF gaps were identified, which are displayed in 5c images. These gaps represent the demineralized areas not infiltrated by resin materials.

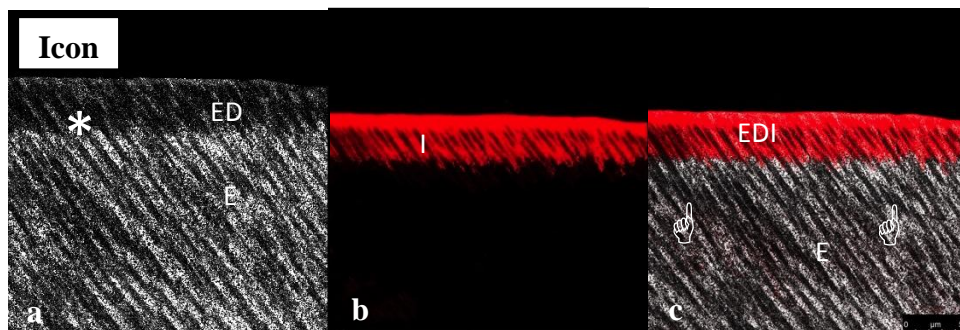


Figure 2. Confocal micrographs showing the penetration of the commercial material Icon. Note the demineralized area (asterisks in a). Sequentially, red areas show the infiltrant impregnated with

rodhamine b into the lesion body (b). The pointers (c) indicate the overlapping of the demoralized areas and red areas, indicating the materials impregnated with rodhamine b into the lesion body. E- sound enamel, ED- enamel area demineralized , I- experimental infiltrant, EDI- enamel area demineralized and infiltrated with experimental infiltrant.

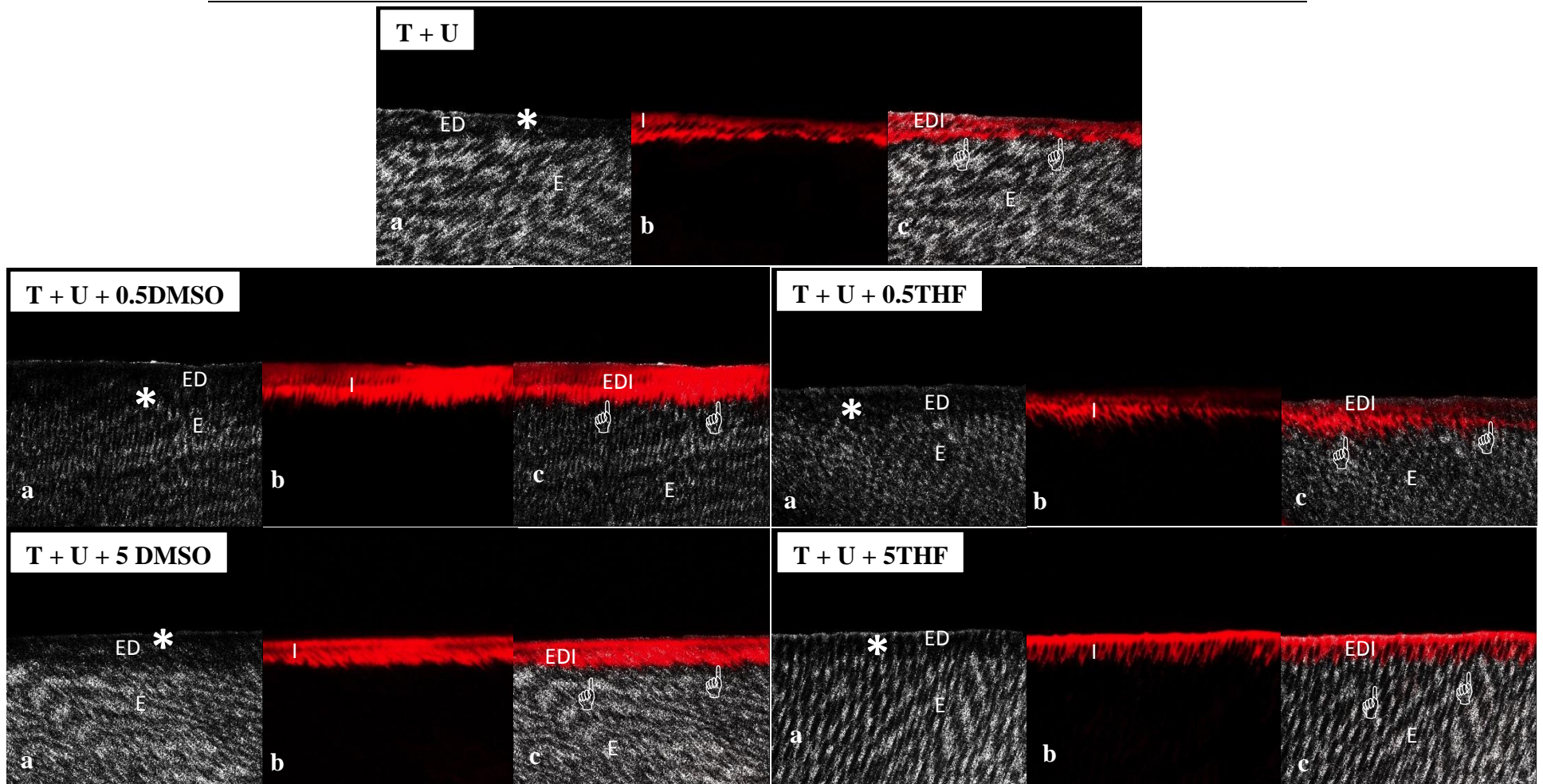


Figure 3. Confocal micrographs showing the penetration of the experimental materials with monomer UDMA. Note the demineralized area (asterisks in a for all groups). Sequentially, red areas show the infiltrants impregnated with rodhamine b into the lesion body (b for all groups). The pointers (c for all groups) indicate the overlapping of the demoralized areas and red areas, indicating the materials impregnated with rodhamine b into the lesion body. E- sound enamel, ED- enamel area demineralized , I- experimental infiltrant, EDI- enamel area demineralized and infiltrated with experimental infiltrant.

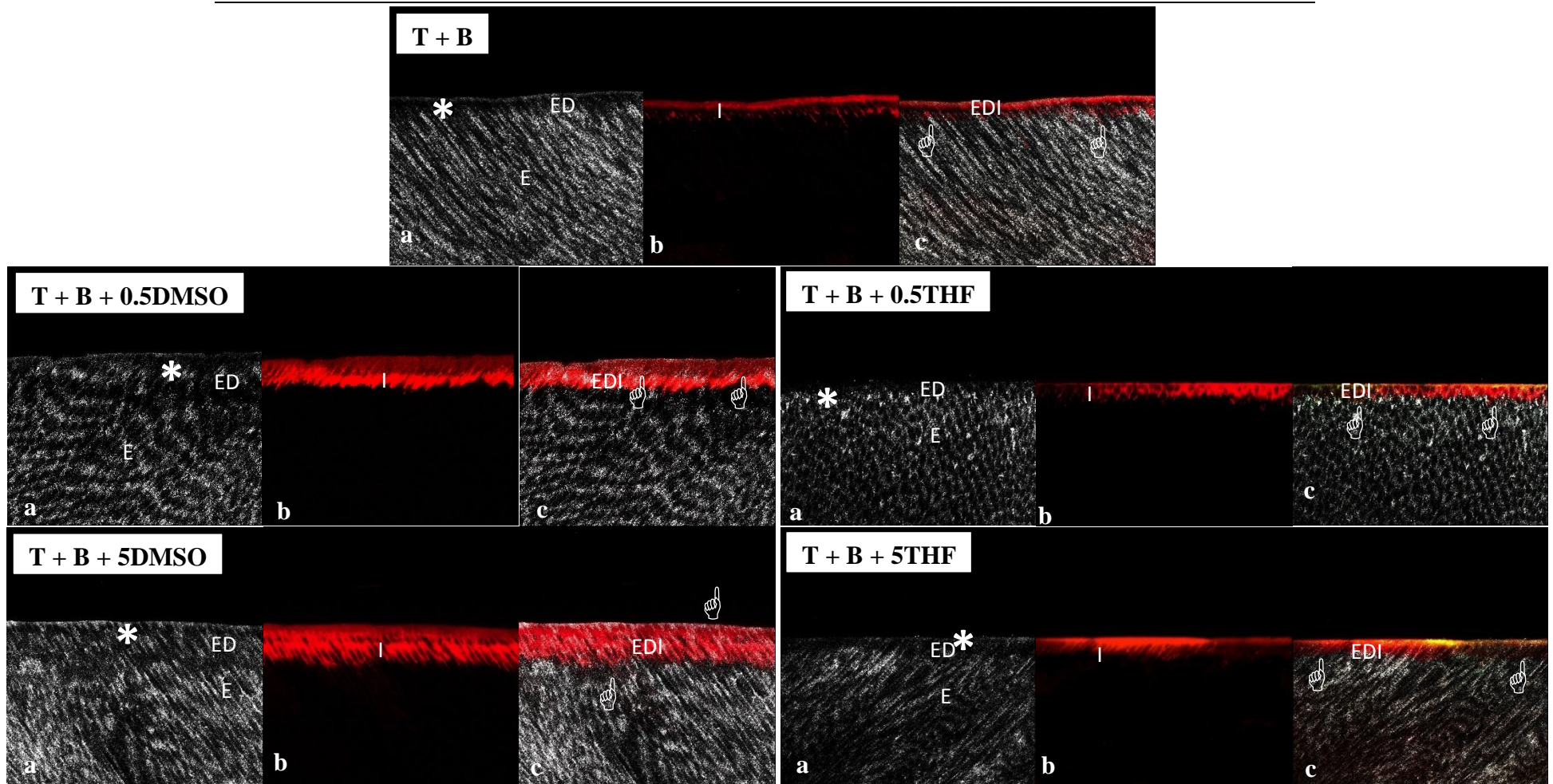


Figure 4. Confocal micrographs showing the penetration of the experimental materials with monomer BisEMA. Note the demineralized area (asterisks in a) for all groups). Sequentially, red areas show the infiltrants impregnated with rodhamine b into the lesion body (b for all groups). The pointers (c for all groups) indicate the overlapping of the demoralized areas and red areas, showing the materials impregnated with rodhamine b into the lesion body. . E- sound enamel, ED- enamel area demineralized , I- experimental infiltrant, EDI- enamel area demineralized and infiltrated with experimental infiltrant.

### DISCUSSION

Nowadays, no restorative material can adequately replace the form, strength, anatomy or appearance of the natural tooth. The cavities minimally invasive reduce the unnecessary destruction of tooth structure and facilitate the use of biomimetic adhesives and restorative materials<sup>22</sup>. Therefore, it is important to develop materials that are appropriate to this new concept of treatment, as with infiltrating resin<sup>23</sup>.

The present results demonstrated that the experimental infiltrants induced differences in the contact angle, Knoop hardness and penetrability into the caries like lesions. Thus, the hypotheses that there would no differences on the Knoop hardness, contact angle and penetrability of infiltrants were rejected. In this study was found influence of the incorporation of dimethyl sulfoxide (DMSO) and tetrahydrofuran (THF) in the physicochemical properties (contact angle and the Knoop hardness) of experimental infiltrants. The results showed the smallest contact angle by Icon<sup>®</sup>, which was significantly lower than the other groups. The contact angle is used to evaluate the ability of the wetness liquids on a given substrate.

On the glass slides, contact angle varied following the viscosity of the functional monomers used in the formulation of the standardized blend<sup>24</sup>. Thereby, the incorporation of solvents THF or DMSO did not influence the viscosity of the experimental infiltrants. The aspect shown in the study was the influence of hydrophobic monomer with higher molecular weight (UDMA and BisEMA). The comparison between these two monomers, BisEMA has higher molecular weight, less flexibility, higher viscosity and higher hydrophobicity<sup>25</sup>. The UDMA has a lower viscosity and lower molecular weight and form more linear polymer chains than BisEMA. However, these differences in characteristics caused no significant change in contact angle of infiltrants even with the incorporation of solvents DMSO and THF. The Icon<sup>®</sup> is composed predominantly of TEGDMA, which presents low molecular weight, high flexibility, high degree of conversion, lower viscosity,<sup>26</sup> which may be responsible for the improved performance of this material compared to the experimental infiltrants.

The contact angle  $\theta$  is a function of both dispersive adhesion and the cohesion within the liquid adhesive. If there is strong adhesion to the substrate surface and weak

cohesion within the liquid, there is a higher degree of wetting, often termed lyophilic conditions. Conversely, a combination of weak adhesion and strong cohesion, referred to as lyophobic conditions, results in higher contact angles and poor wetting of the substrate surface, that is droplets form on the surface rather than a film of fluid<sup>27</sup>. These relationships can be put in this study in another way. When test the Icon®, the forces between glass slide and liquid are greater than the cohesive forces within the infiltrant, and the liquid can spread over the glass slide surface. This is due the Icon® to be the basis TEGDMA, low monomer viscosity, lower molecular weight and higher concentration of double bonds. It is a relatively flexible linear molecule, allowing greater crosslink density in the polymer, with a more heterogeneous structure, with higher water sorption<sup>28</sup>. Therefore, a better wetting can be achieved by lowering the surface tension of infiltrant. In the experimental infiltrants, the cohesion within the infiltrant may have exceeded the surface energy of the glass slide, such as poor wetting was noted. Although experimental infiltrants present in its composition hydrophilic solvents DMSO and THF, the monomers UDMA and BisEMA are highly hydrophobic<sup>25,28</sup>. Thus, the higher contact angle of the experimental materials was noted.

The results of this study corroborate with previous study<sup>8</sup> considered an attempt to add solvents ethanol and HEMA in the experimental infiltrants to reduce viscosity and increase the depth of penetration; but the results of this study indicated that adding a solvent to infiltrant blends, except for UDMA blends, damaged the KHN. However, the addition of solvents/diluents did not show a difference among the groups in homogeneity of penetration.

The blends T + U + 0.5% DMSO and T + U + 5% THF resulted in an increased KHN. Thus, the addition of UDMA to TEGDMA could improve the mechanical properties of infiltrants<sup>8,29</sup>. The monomer UDMA shows lower viscosity than BisEMA, which comparatively could improve the penetration coefficient, there is also more flexible urethane bonding, which can improve the mechanical properties of materials using this monomer<sup>30</sup>, especially when compared to TEGDMA/BISEMA groups. The blends T + B; T + B + 5% DMSO and T + B + 0.5% THF showed the lowest values of KHN.

The H-bonding capacity of a solvent has been shown to be an important property in the removal of water<sup>31,32</sup>. However, this feature can also facilitate the attraction between polymer and solvent, increasing the retention of solvent and water<sup>33,34</sup>. The solvents tested (DMSO and THF), are aprotic solvents. However, the solvent THF having a highly volatile character (143 mmHg of vapor pressure), demonstrated a good water-removing capacity and enhanced evaporation<sup>15</sup>, which may explain the better results of KHN were added when higher percentages of THF in the experimental infiltrants association with the UDMA. In turn, DMSO has a low vapor pressure (0.42mmHg)<sup>35</sup>, which hinders its evaporation resulting in structures with a residual solvent, showing lower performance KHN when use higher percentage (5%) regardless of the monomer used. In association with UDMA to a lesser percentage (T + U + 0.5% DMSO), it may not have influenced the properties of the experimental infiltrant, showing high values of KHN similar to Icon<sup>®</sup>. It was observed that the greater the depth, the higher values of KHN. This can be explained because hardness is an indirect method for evaluating demineralization<sup>36,37</sup>. Therefore, may be areas of sound enamel.

According to the CLSM images analysis, variation in the blend composition through the addition of solvents showed a good penetration into the demineralize enamel, lesion except for groups T + B; T + B + 0.5% THF and T + B + 0.5% DMSO. The monomer UDMA shows lower viscosity than BisEMA<sup>30</sup>, which could improve the penetrability of the materials based on UDMA. The association of monomer BISEMA with low percentage of solvents (0.5% THF or 0.5% DMSO) might not be sufficient to reduce the viscosity of infiltrants and promote good penetration into the demineralized area.

The development of new formulations is a promising method for a significant improvement of the performance of these materials, increasing the penetration and establishment of new approaches to an interception of ECLL. Further studies should be conducted to evaluate the effect of incorporation of solvents in experimental infiltrants the contact angle with dental structure (enamel), since this study was conducted in glass slice, which varied the contact angle as a function of monomers used.

### **CONCLUSION**

The results of this study showed that the addition of low concentrations of DMSO and THF in the experimental infiltrants increased KHN. The blends with solvents in lower concentration (0.5%) associate with monomer BisEMA not promoted good penetration of the experimental infiltrants. In addition, Icon showed the lowest contact angle. There was reduction on contact angle only when high concentration of DMSO (5%) was added to the TEGDMA and UDMA infiltrant.

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### CAPÍTULO 2

#### **Influence of alternative solvents on the degree of conversion, bond strength and mechanical properties of experimental resin infiltrants**

##### **ABSTRACT**

**Objective:** To assess the influence of monomers and solvents (tetrahydrofuran (THF) and dimethyl sulfoxide (DMSO)) on degree of conversion, microtensile bond strength and mechanical properties of experimental resin infiltrants. **Methods:** Eleven groups were evaluated: Icon; 75% TEGDMA (T) + 25%UDMA (U); 75%T + 25%U + 0,5%DMSO; 75% T + 25% U + 5% DMSO; 75% T + 25% U + 0,5% THF; 75% T + 25% U + 5% THF; 75% T + 25% BisEMA(B) ; 75% T + 25% B + 0,5% DMSO; 75% T + 25% B + 5% DMSO; 75% T + 25% B + 0,5% THF; 75% T + 25% B + 5% THF. Degree of conversion, tensile cohesive strength, flexural strength and elastic modulus of resins infiltrants were evaluated. Microtensile bond strength was evaluated on bovine teeth after infiltration of resin infiltrants. Data were submitted to one-way ANOVA and Tukey's test ( $\alpha = 0.05$ ). **Results:** The degree of conversion means varied from the lowest for T+B+5%THF (41.9%) to the highest for T + U + 5%THF (62.1%). The flexural strength and E-modulus varied from the lowest for T + U + 5% DMSO (18.5MPa and 9.7 GPa, respectively) to the highest for T + B (96.5 MPa and 0.49 GPa, respectively). Icon showed the highest bond strength (19.3 MPa) and cohesive strength (62.2 MPa), whereas T+U+5%DMSO (9.7 MPa) and T + B + 5% DMSO (9.8 MPa) the lowest bond strength; T + B + 0.5% DMSO (12.3 MPa) showed the lowest cohesive strength. **Significance:** Addition of DMSO and THF didn't improve the bond strength to demineralized enamel and jeopardized the mechanical properties of resins infiltrants. The 0.5% DMSO produced few deleterious effects on mechanical properties of resin infiltrants.

**Key-words:** degree of conversion, bond strength, cohesive strength, flexural strength, modulus of elasticity, solvent, infiltrant.

### INTRODUCTION

The white spot lesions are caused by demineralization of enamel. White spot prevalence of 50% (Gorelick *et al.*, 1982), 60% (Hadler-Oslen *et al.*, 2011), or even 97% (Boersma *et al.*, 2005) after bonded or banded orthodontic treatment has been reported, affecting especially labial surfaces of maxillary incisors.

Remineralization using fluoride or casein phosphopeptide amorphous calcium phosphate has been shown to positively influence caries arrest (Baily *et al.*, 2009; Willmot, 2004). However, deeper lesions do not remineralise completely, as the formation of a hyperremineralised surface layer hampers the remineralisation of the subsurface lesion body (Cate *et al.*, 1977; Naumova *et al.*, 2012). Fluoride topical application with different concentrations have been long used to arrest and remineralize advanced eroded enamel lesions; however, there are still contradictory results regarding the capability of the fluoride to remineralize the whole depth of the subsurface decalcified enamel lesions and the time needed to completely remineralize these lesions (Reynolds *et al.*, 2008).

A technique of applying a low-viscosity resin have been studied and presented successful results (Paris *et al.*, 2010; Meyer-Lueckel *et al.*, 2012). The aim of this technique is to occlude the highly porous structures of incipient enamel lesions (Davilla *et al.*, 1975; Robinson *et al.*, 1976). The infiltration term seems most appropriate to describe the treatment approach using low-viscosity resin mixtures with high penetration capabilities on subsurface enamel lesions (Kielbassa *et al.*, 2009).

The current results of the material Icon<sup>®</sup> (DMG, Hamburg, Germany) to caries infiltration is an efficacious method to hamper progression of non cavitated proximal lesions extending radiographically into the inner half of enamel up to the outer third of dentin after a period of 3 years (Meyer-Lueckel *et al.*, 2012). Icon<sup>®</sup> is based on triethyleneglycol dimethacrylate (TEGDMA) that present potentially high water sorption over time. Others hydrophobic monomer combinations such as urethane dimetrachrylate (UDMA) and ethoxylated bisphenol A glycidyl dimethacrylate (BisEMA) should be evaluated to increase the water stability of resins infiltrants.

In order to increase the therapeutic effect in caries lesion, ideally an infiltrant should present low viscosity, low surface tension and acceptable mechanical properties that support dental abrasion and oral degradation (Paris *et al.*, 2007). Introducing a solvent with advantageous volatile properties may improve the stability of the polymer (Fontes *et al.*, 2013) due to its volatile properties (e.g., high vapor pressure). Tetrahydrofuran (THF) was recently reported as a feasible alternative solvent for a model adhesive system (Fontes *et al.*, 2009). Similarly, dimethyl sulfoxide (DMSO) is a solvent used extensively in many types of chemical reactions requiring high polarity as well as analytical methods and practical applications of biochemistry and pharmacology (Vaisman and Berkowitz, 1992; Chalaris *et al.* 2008).

The addition of DMSO or THF into resin infiltrants could increase the wettability, penetration and imbrication into demineralized enamel, being beneficial to occlude caries lesion. Thus, the aim of this study is to assess the influence of solvents on degree of conversion, bond strength, cohesive strength and modulus of elasticity) of experimental resin infiltrants. The null hypothesis tested was that different solvents and monomers would not attain differences on the physicochemical properties of infiltrants.

## MATERIAL AND METHODS

### *1. Infiltrant preparation*

The following monomers were used: ethoxylated bisphenol A glycidyl dimethacrylate (BisEMA) (Sigma-Aldrich Inc., Batch #03514HF), urethane dimethacrylate (UDMA) (Sigma-Aldrich Inc., Batch #09405B) and triethyleneglycol dimethacrylate (TEGDMA) (Sigma-Aldrich Inc., Batch #01612M). The solvents dimethyl sulfoxide (DMSO) and tetrahydrofuran (THF) (Sigma-Aldrich Inc., Batch #51496AM) were in different ratios, as described in Table 1.

The monomers resin blend were mixed up in brown glass jars, for each experimental group. For all blends, the photoinitiator system selected was camphorquinone (CQ) (Sigma-Aldrich, Batch #532604) and dimethyl aminoethyl methacrylate (DMAEMA) (Sigma-Aldrich Inc., Batch #BCBF8391V) as co-initiator (proportion 1:2 by weight). The

inhibitor butylated hydroxytoluene (BHT) (Sigma-Aldrich Inc., Batch #04416KD) was added to the resin blends with a concentration of 0.1 wt% in order to avoid the spontaneous polymerization of the monomers. Also, the light-curing initiator system was thoroughly dissolved in the monomer matrix with the concentration of 1.5 wt% (0.5% CQ/ 1% DMAEMA). In order to avoid premature polymerization, the resin blend groups were stored at 4°C until use.

Table 1-Infiltrant blends composition.

Infiltrant	Composition (wt,%)
Icon®	Methacrylate resin
T + B	75% TEGDMA+ 25% BisEMA
T + B + 0.5DMSO	T+B+0.5%DMSO
T + B + 5DMSO	T+B+5%DMSO
T + B + 0.5 THF	T+B +0.5%THF
T+B+5 THF	T+B+5%THF
T + U	75% TEGDMA +25%UDMA
T + U + 0.5 DMSO	T+U+0.5%DMSO
T + U + 5 DMSO	T+U +5%DMSO
T + U + 0.5 THF	T+U +0.5%THF
T + U + 5 THF	T+U +5%THF

T- triethyleneglycol dimethacrylate; B- ethoxylated bisphenol A glycidyl dimethacrylate; U- urethane dimethacrylate; DMSO- Dimethyl Sulfoxide; THF-tetrahydrofuran.

## 2. Microtensile bond strength

### 2.1.Preparation of enamel blocks

One hundred and ten bovine mandibular incisors without cracks or caries were collected. The teeth were cleaned and stored in 0.1% thymol solution up to 1 month after extraction. The enamel surfaces were flattened (5 x 5 mm) on a water-cooled mechanical grinding machine using 340- and 600-grit Al<sub>2</sub>O<sub>3</sub> abrasive paper (Aropol E, Arotec S.A,

Ind.&Com., São Paulo, Brazil). The roots were cut 1 mm below the cement enamel junction using a diamond disc (Isomet, Buehler Ltd., Lake Bluff, IL, USA) and discarded. The teeth crowns were cut using a diamond disc (Isomet, Buehler Ltd, Lake Bluff, IL, USA.) to obtain blocks of enamel (5x5mm).

### **2.2. Artificial enamel caries-like lesion formation**

The subsurface enamel caries-like lesions (ECLL) were produced in the sound enamel surface. Each enamel block was covered with double coats of acid-resistant nail varnish (Colorama®, São Paulo, Brazil) except to the polished enamel area (5 x 5 mm). The ECLL was produced by immersion of each enamel surface into 50 mL of a demineralizing solution containing 0.05 M acetate buffer 50% hydroxyapatite saturated from enamel powder, pH 5.0, for 16 h at 37 °C (Paes *et al.*, 2003). To prepare the solution, enamel powder (particles of 74-105 µm) was agitated into 0.05 sodium acetate buffer, pH 5.0, for 96 h at 37°C (0.50 g/L) (Gwinnett *et al.*, 1967; Jacobsen *et al.*, 1995). The solution was used in a ratio of 2.0 mL/mm<sup>2</sup> of exposed enamel area. Immersion period of 16 h was determined in a previous study, by analyzing thin enamel slices with polarized light microscopy. It was clearly observed the presence of subsurface ECLL. Calcium concentration into the solution was 66.3 µg/mL, which was determined by atomic absorption spectrometry with flame spectrophotometer model 506 (Perkin Elmer); phosphorus concentration was about 32 µg/mL, which was determined by colorimetric method of Fiske and Subbarow (1925) with spectrophotometer model 800M (Analyzer) adjusted at 660 nm.

### **2.3. Specimen preparation**

The enamel blocks with ECLL were randomly distributed into eleven groups (Table 1) (n = 10) according to the composition of low viscosity resin materials. The previously determined area (5x5mm) on enamel surface blocks was etched with 37% phosphoric acid gel (Vigodent, Rio de Janeiro, RJ, Brazil) for 60 s (Meyer-Lueckel *et al.*, 2007), rinsed for 10 s and dried with compressed air for 15s. The experimental infiltrants



were applied using micro-brush for 60 s (Meyer-Lueckel *et al.*, 2006) in order to improve the penetration into the etched enamel. Block surface was air-dried for 15 s in order to evaporate the solvent. Infiltrants were then light cured for 60 s using Ultralume 5 (Ultradent, South Jordan, UT, USA) with 1000 mW/cm<sup>2</sup>. Blocks of experimental infiltrants with 4 mm height (increments of 2 mm-thickness) were done using a silicon mold, individually light cured for 60 s. Then, enamel blocks were stored in 100 % humidity for 24 hours at 37°C. Afterwards, each enamel block was longitudinally cut into slices of 1 mm thickness, using water-cooled diamond blade (Isomet, Buehler Ltd, Lake Bluff, IL, USA). Four slices were obtained of each block and trimmed to a dumbbell shape using a cylindrical diamond bur (FF 1092, KG Sorensen, São Paulo, SP, Brazil) in a high-speed hand piece.

#### **2.4. Microtensile bond strength test ( $\mu$ TBS)**

The dumbbell-shaped specimens were individually fixed to apparatus and tested using a universal testing machine (EZ-TEST) with a 50 N load at 0.5 mm/min cross head speed. The cross-sectional area at the site of fracture was measured with a digital caliper (Starrett 727, Starrett Indústria e Comércio LTDA, Itu, SP, Brazil) with an accuracy of 0.01mm. The microtensile bond strength was calculated in MPa, according to the following formula:  $R = F \times 0,098/A$ , where A= Bonding surface area in cm<sup>2</sup>, F = value of force obtained during the test in kgf, R = bond strength in MPa. Data were submitted to one-way ANOVA and Tukey's test ( $\alpha = 0.05$ ).

#### **2.5. Analysis of failure pattern**

The fractured specimens were fixed on metallic stubs with double-sided carbon tape (Electron Microscopy Sciences, Washington, USA). The stubs with the fractured specimens were ultrasonically cleaned with distilled water for 10 minutes and dehumidified for 2 hours in an oven (Fanem, Kiln Drying and Sterilization - Model 315 SE, SP, Brazil) at 40°C for 6 hours. All specimens were gold-sputter coated (Balzers model SCD 050 sputter coater, Balzers Union Aktiengesellschaft, Liechtenstein Fürstentum, Germany) at 40 mA

for 120 s and observed in Scanning Electron Microscope (SEM) at an accelerating voltage of 15 kV, working distance (WD) of 33 mm, spot size 44 and magnification of X50 and X200. The failure patterns were classified as: Type-I cohesive failure in infiltrant; Type II – cohesive failure in enamel; Type III - mixed failure between the infiltrant layer and the enamel. A blind calibrated examiner evaluated the failure pattern. The intra-examiner coincidence level was analyzed by Spearman's correlation and was observed 95% of coincidence, obtained in a 7 days interval evaluation.

### ***3. Flexural strength and E-modulus (EM)***

Bar specimens (9 mm length, 2 mm thick and 2 mm width) were produced in metallic molds. The specimens (n = 10) were light-cured at 1000 mW/cm<sup>2</sup> at 3 different points each for 60 s using Ultralume 5 (Ultradent, South Jordan, Utah, USA) under a polyester strip (Airon Maquira Dental Products Industry, Maringa, Brazil). Specimens were dry stored for 24 h in lightproof containers at 37°C.

To assess the flexural strength and elastic modulus the three point-bending test was performed in a universal testing machine (INSTRON, model 4111, Instron Corp., OH, USA). The test was performed with a crosshead speed of 0.5 mm/min and a cell load of 50 N until fracture. The distance between supports was 3 mm; EM was calculated using Bluehill 2 software (Illinois Tool Works Inc., IL, USA) coupled to the universal testing machine. Data were submitted to one-way ANOVA and Tukey's test ( $\alpha = 0.05$ ).

### ***4. Degree conversion***

The degree of conversion (DC) of the resin infiltrants commercial was evaluated using the Attenuated Total Reflection Fourier-Transform Infrared spectrophotometer (Nicolet 5700, Thermo Fisher Scientific, Loughborough, UK) equipped with an ATR crystal. The material was dropped on the base of FTIR (n = 3) and heights of peaks were collected according to the aliphatic and aromatic peaks.

The same drop (n = 3) was light-cured (60 s; 1000 mW/cm<sup>2</sup>, Optilux VLC, Demetron Kerr, Orange, USA) and new heights of peaks were collected. The remaining unconverted double bonds were determined by comparing the ratio of the aliphatic C=C

absorption peak at  $1638\text{ cm}^{-1}$  to the aromatic group C = C peak at  $1608\text{ cm}^{-1}$  between the polymerized and unpolymerized specimens (Atai *et al.*, 2004). Data were submitted to one-way ANOVA and Tukey's test ( $\alpha=0.05$ ).

### 5. Tensile Cohesive Strength

The resin infiltrants ( $n = 10$ ) were inserted into silicon molds in order to obtain dumbbell shape (2 mm height x 8 mm length x 1.5 mm constriction region) specimens. The material surface was covered with a Mylar strip and then light cured using Ultralume 5 (Ultradent, South Jordan, Utah, USA). The dumbbell shaped specimens were removed of the molds and polished under irrigation with 600-1200 grit silicon carbide (SiC) abrasive paper. After storage for 24 h, specimens were tested until the failure under tensile using an universal testing machine (EZ Test) with a 50N load at 0.5 mm/min crosshead speed. The exact area of the transversal section of the fractured specimens was measured with a digital caliper. The tensile cohesive strength was calculated in MPa. Data were submitted to one-way ANOVA and Tukey's test ( $\alpha=0.05$ ).

## RESULTS

The outcomes (means and standard deviations) of the degree conversion, flexural strength, E-modulus, microtensile bond strength and tensile cohesive strength are presented in Table 2.

Icon<sup>®</sup> showed the highest bond strength (19.3MPa), statistically similar to T + U + 0.5% DMSO (16.2MPa). Intermediate values are presented by the groups (T + U + 0.5% THF; T + U; T + B; T + B + 0.5% DMSO; T + U + 5% THF; T + B + 0.5% THF; T + B + 5% THF). The addition of 5% DMSO into resin infiltrants significantly decrease the microtensile bond strength.

The flexural strength and E-modulus varied from the lowest for T + U + 5% DMSO (18.5 MPa and 9.7GPa, respectively) to the highest for T + B (96.5MPa and 0.49 GPa, respectively). The solvent-free experimental infiltrants (T + B and T + U) showed statistically higher flexural strength than other groups. The addition of DMSO on T + U blends significantly decrease flexural strength compared to THF. For T + B blends there

was no significant differences between DMSO and THF. For EM, the ranking was as follows:  $T + B \geq T + B + 0.5\%THF \geq T + U + 0.5\%THF \geq T + U \geq T + B + 0.5\%DMSO = T + U + 5\%THF \geq T + B + 5\%THF = T + B + 5\%DMSO \geq Icon^{\circledR} \geq T + U + 0.5\%DMSO > T + U + 5\% DMSO$ .

The lowest DC varied from the lowest  $T + B + 5\% THF$  (41.9%) to the highest  $T + U + 5\% THF$  (62.1%). The blends containing UDMA showed statistically higher degree of conversion than BisEMA. Icon presented intermediary outcomes (50.4%), significantly higher than  $T + B + 5\% DMSO$  (42.8%),  $T + B + 0.5\% THF$  (42.1%), and  $T + B + 5\% THF$  (41.9%).

Figure 1 shows the graphic illustration of the distribution of fracture patterns observed for each experimental group. The groups  $T + U$ ,  $T + U + 0.5THF$ ,  $T + B$ ,  $T + B + 0.5\%DMSO$  and  $T + B + 0.5\%THF$  presented mixed predominant fracture pattern. The groups  $T + U + 5\% DMSO$ ,  $T + U + 5\%THF$   $T + B + 5\%DMSO$  and  $T + B + 5\%THF$  showed predominant cohesive failures in enamel. Groups that had the predominant cohesive in infiltrant are  $T + U + 0,5\%DMSO$  and  $Icon^{\circledR}$ .

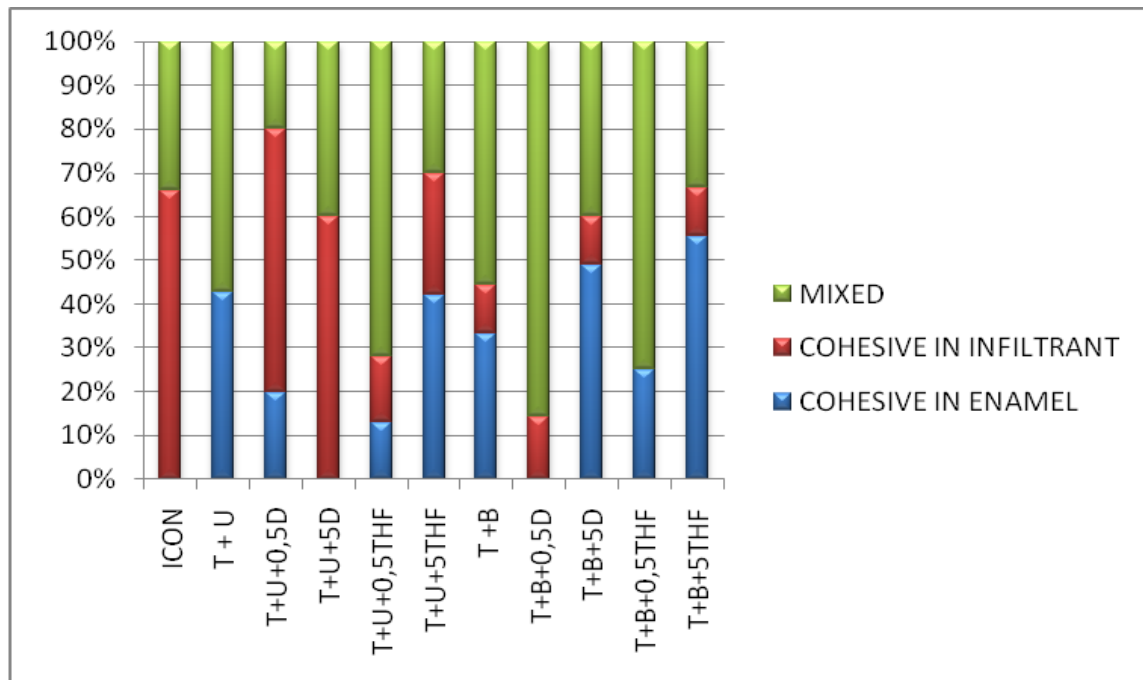


Figure1. Failure pattern distribution of different groups tested.

Table 2- Mean (standard deviation) values of degree of conversion, flexural strength, elastic modulus, bond strength and tensile cohesive strength of resin infiltrants,

Groups	Degree of conversion (%)	Flexural strength (MPa)	Elastic Modulus (GPa)	Bond Strength (MPa)	Tensile cohesive strength (MPa)
Icon®	50.4 (1.6) b	62.4 (20.3) bc	0.28 (0.10) de	19.3 (1.4) a	62.2 (2.0) a
T+B	46.8 (1.9) bcd	96.5 (20.9) a	0.49 (0.10) a	14.3 (1.4) b	28.3 (6.6) cd
T+B+0.5DMSO	47.9 (1.4) bc	64.8 (15.6) bc	0.35 (0.12) bcde	14.1 (1.2) b	12.3 (1.9) e
T+B+5DMSO	42.8 (0.8) cd	64.6 (4.9) bc	0.28 (0.05) cde	9.8 (1.9) c	22.6 (0.9) d
T+B+0.5THF	42.1 (3.2) d	85.8 (14.4) b	0.48 (0.10) ab	13.8 (0.7) b	39.6 (4.1) bc
T+B+5THF	41.9 (3.9) d	70.9 (17.8) b	0.32 (0.04) cde	13.6 (0.8) b	30.0 (5.1) bcd
T+U	61.2 (0.4) a	96.1 (20.4) a	0.39 (0.10) abcd	14.4 (1.8) b	30.0 (5.1) bcd
T+U+0.5DMSO	61.9 (0.2) a	45.7 (10.6) c	0.25 (0.07) e	16.2 (1.4) ab	37.6 (15.2) bc
T+U+5DMSO	61.5 (0.2) a	18.5 (6.2) d	0.07 (0.02) f	9.7 (1.8) c	38.2 (7.3) bc
T+U+0.5THF	61.2 (0.2) a	83.7 (18.5) b	0.41 (0.08) abc	16.1 (1.6) b	40.9 (2.9) bc
T+U+5THF	62.1 (0.3) a	61.2 (17.8) bc	0.34 (0.10) bcde	14.0 (0.9) b	42.3 (4.0) b

Different letters in columns indicate statistically significant difference ( $p < 0.05$ ). T- triethyleneglycol dimethacrylate; B- ethoxylated bisphenol A glycidyl dimethacrylate; U- urethane dimethacrylate (UDMA); DMSO- dimethyl sulfoxide; THF- tetrahydrofuran.

### DISCUSSION

In order to improve the properties of infiltrants, in this study other monomer combinations, incorporated with different solvents, were studied to increase the longevity of these materials. The null hypothesis of this study was rejected, because the experimental infiltrants composed by different solvents and monomers showed differences on the bond strength, cohesive strength, flexural strength and e-modulus. The exception was degree of conversion, in which there was no significant difference between different monomers and solvents.

According to the results of this study, the blends composed of BisEMA and TEGDMA showed significantly lower degree of conversion than UDMA and TEGDMA infiltrants. These results corroborate prior study (Araújo *et al.*, 2013). This may be related to the high molecular weight (540 g/mol) of BisEMA and its lower chain flexibility than TEGDMA or UDMA (Sideridou *et al.*, 2002). However, the BisEMA increases the hydrophobic characteristics of the infiltrant, which could be interesting because more hydrophobic material tends to show reduced degradation in the oral environment (Araújo, *et al.*, 2013). The UDMA, as well TEGDMA, presents linear chains. UDMA presents flexible chains and two aliphatic urethane linkages that are capable of forming hydrogen bonds. UDMA has a small size, low viscosity (1.23 Pa.s), and a high amount of double bonds (Araújo *et al.*, 2013). Monomer conversion after polymerization of light-cured materials affects various mechanical properties, such as the tensile, compressive, and flexural strengths, elastic modulus, wear, and hardness (Asmussen, 1982; Ferracane, 1985).

The addition of DMSO or THF did not significantly affect the DC, independent the amount or type of solvent added to the blend. The DC is affected by the monomers composed of the blends. The results were statistically similar groups contained within the same monomer. However, polymers that have similar degree of conversion (DC) values can present different crosslink material strengths, since the mechanical properties are associated with the polymeric structure (Ye *et al.*, 2007a).

In the current study the results showed differences in mechanical properties when solvent DMSO or THF was added in the infiltrants. This can be shown in the results

of bond strength, e-modulus and flexural strength. In all groups when the percentage of solvent increased from 0.5% to 5% decreased the properties mentioned, for both DMSO and THF. Despite increasing the DC, these results demonstrate that the chemical complexity associated with presence of solvent in experimental infiltrant caused substantial differences in mechanical properties (Gaglianone *et al.*, 2012).

The addition of higher concentration of solvents (5wt%) to the infiltrants caused an increase on cohesive failures in enamel. The higher solvent content may impair the penetration of infiltrant into the enamel caries lesion, as a function of a possible reduction in penetration coefficient. The infiltrants should penetrate several hundreds of micrometers into the enamel (Paris *et al.*, 2012). Thus, solvents could not be completely eliminated by evaporation before light curing, and polymerization may be compromised (Yiu *et al.*, 2005; Ye *et al.*, 2007b; Pashley *et al.*, 2011) damaging the mechanical properties.

The group T + U + 0.5% DMSO and Icon<sup>®</sup> showed the highest bond strength values and pattern failure cohesive in infiltrant. The monomer UDMA shows lower viscosity than BisEMA, which comparatively could improve the penetration coefficient. Besides, there is also more flexible urethane bonding, which can improve the bond strength of materials using this monomer (Glenn, 1982).

The solvents decreased the e-modulus and flexural strength of experimental infiltrants. These results corroborate other studies indicating that the e-modulus decreased with an increase in solvent content, which suggests changes in the polymer crosslinking (Ye *et al.*, 2007a, Gaglianone *et al.*, 2012). Although some standard tests suggested changes properties, the incorporation of an alternative solvent in infiltrants composition is a promising method for significantly improving the performance of these materials. Studies about stability of resin infiltrants with addition of DMSO and THF could clarify the role of these components on longevity of the caries like lesions infiltrated with resin materials.

## CONCLUSION

Based on the results of the present in vitro study, it can be concluded that addition of DMSO and THF did not impair the bond strength, but reduced the mechanical

properties of resin infiltrants. The monomers affected the DC. Best results were shown by UDMA and TEGDMA blends. The addition of 0.5% of solvents (THF or DMSO) showed the lowest deleterious effects on the mechanical properties of resin infiltrants compared to higher percentage of solvents (5%).

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### **CONSIDERAÇÕES GERAIS**

Uma terapia alternativa promissora para tratar lesões iniciais de cárie pode ser a infiltração com resinas de baixa viscosidade (Gelani *et al.*, 2014). No entanto, em contraste como selamento convencional da cárie, com essa técnica é criada uma barreira de difusão no interior da lesão e não somente superficial, obliterando os poros da lesão e reforçando a estrutura desmineralizada. O material introduzido no mercado com essa concepção foi o Icon<sup>®</sup>. Este material é composto com base no TEGDMA, tem sido estudado e apresentado êxito. Paris *et al.* (2010b) após 18 meses obtiveram resultados satisfatórios, demonstrando que a progressão das lesões de cárie interproximal *in vivo* foi interrompida. Entretanto, devido às características de sorção de água apresentada pelo TEGDMA, estudo de outras combinações monoméricas para aumentar a longevidade destes materiais, passou a ser considerado.

No capítulo 1 foi verificado que a incorporação dos solventes não melhorou o molhamento dos materiais, apresentando maiores ângulo de contato comparados com o Icon<sup>®</sup>. Foi verificado, como em estudo anterior, que o ângulo de contato na placa de vidro varia em função da viscosidade dos monômeros funcionais usados na formulação (Feitosa *et al.* 2014). Ao comparar estes monômeros, o BisEMA tem maior peso molecular, menos flexibilidade, maior viscosidade e hidrofobicidade superior (Sideridou *et al.*, 2002). O UDMA tem menor viscosidade, peso molecular mais baixo e forma cadeias poliméricas mais lineares que o BisEMA. O Icon<sup>®</sup> é composto predominantemente de TEGDMA, que apresenta baixo peso molecular, alta flexibilidade, alto grau de conversão, de baixa viscosidade (Meyer Lückel and Paris 2008), que podem ser responsáveis pelo melhor desempenho deste material em comparação com os infiltrantes experimentais. O ângulo de contato é uma função tanto da adesão dispersiva e coesão dentro do adesivo líquido. Se houver forte adesão à superfície do substrato e fraca coesão no interior do líquido, existe elevado grau de molhamento, geralmente conhecidas como condições liofílicas. Por outro lado, uma combinação de fraca adesão e coesão forte, que se refere a condição liofóbica, resulta em elevados ângulos de contato e molhamento insuficiente da superfície do substrato, isto é, formam-se gotas sobre a superfície em vez de uma película de fluido

## **Considerações gerais**

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(Fraunhofer, 2012). Nos infiltrantes experimentais, a coesão do infiltrante pode ter excedido a adesão entre a placa de vidro e o líquido de tal modo que existe um molhamento insuficiente. Embora exista solventes hidrófilos (DMSO e THF) presentes na composição dos infiltrantes experimentais os monômeros utilizados (UDMA e BisEMA) são altamente hidrófobos (Sideridou *et al.*, 2002; Barszczewska-Rybarek, 2009). Assim, pode ser justificado o ângulo de contacto elevado dos materiais.

Ainda no capítulo 1 , também foi verificado através da análise das imagens do confocal que os infiltrantes tiveram uma boa penetração em toda extensão da lesão, exceto as blendas que continham menor porcentagem dos solventes (0,5%) associados com o monômero BisEMA ( T + B + 0.5% DMSO, T + B + 0.5% THF) e a blenda livre de solvente (T + B).

No capítulo 2 os resultados mostraram que o grau de conversão variou em função do monômero presente nos infiltrantes experimentais. As blendas que continham o monômero BisEMA apresentaram ou os menores valores de grau de conversão, e quando o monômero era o UDMA os valores aumentaram. Esse resultado vai de acordo com estudos anteriores (Araujo *et al.*, 2013). O monômero BisEMA apresenta um peso molecular alto com menor flexibilidade que os monômeros TEGDMA e UDMA que apresentam cadeias lineares com maior flexibilidade e conseqüentemente maior grau de conversão (Sideridou *et al.*, 2002; Atai *et al.*, 2005; Araújo *et al.*, 2013). Apesar do aumento do grau de conversão, os resultados demonstram que a complexidade química associada com a presença de solvente no infiltrante experimental causou diferenças substanciais nas propriedades mecânicas(Gaglionone *et al.*, 2012). Isso pode ser observado nos resultados de resistência de união, resistência a flexão e módulo de elasticidade no capítulo 2 e na dureza Knoop no capítulo 1, onde as maiores concentrações dos solventes em combinação com ambos monômeros (UDMA E BisEMA) apresentaram valores menores, exceto para o grupo T+U+5%THF, que pode estar relacionado ao fato de que o UDMA associado ao TEGDMA pode promover melhora nas propriedades mecânicas dos infiltrantes (Atai *et al.*, 2005; Araújo *et al.* 2013). Além disso, o solvente THF possui alta volatilização, demonstrando boa capacidade de remoção da água (Fontes *et al.*, 2013), o que pode explicar os valores altos de dureza Knoop.

## **Considerações gerais**

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A adição de uma maior porcentagem dos solventes (5%) com associação em ambos monômeros UDMA e BisEMA, mostrou maiores falhas coesivas no esmalte no teste de microtração. Isso pode ser explicado por uma melhora na força coesiva dos materiais e talvez, alguma dificuldade na penetração na lesão de cárie de esmalte, como função de possível redução no coeficiente de penetração. Isso também pode ser explicado devido os infiltrantes penetrarem centenas de micrômetros no esmalte (Paris *et al.*, 2012). Nestas profundidades, o solvente é difícil de ser completamente eliminado por evaporação antes da polimerização, comprometendo o polímero (Yiu *et al.*, 2005; Ye *et al.*, 2007b; Pashley *et al.*, 2011) e danificando as propriedades mecânicas. Portanto, seria recomendado aumentar o tempo de evaporação além de 15 segundos.

O grupo T + U + 0,5% DMSO assim como Icon® apresentaram os maiores valores de resistência de união e padrão de fratura coesiva no infiltrante. O monômero UDMA apresenta menor viscosidade do que BisEMA, que comparativamente poderia melhorar o coeficiente de penetração, também há ligação uretano mais flexível, que pode melhorar a resistência de união de materiais utilizando este monômero (Glenn, 1982).

Neste estudo, o módulo de elasticidade e a resistência à flexão de infiltrantes experimentais com solvente não foram reduzidos. Estes resultados estão de acordo com outros estudos que indicam que o módulo de elasticidade diminui com o aumento do teor de solvente, o que sugere que as alterações na ligação cruzada do polímero (Ye *et al.*, 2007a, Gagliane 2012).

Novos estudos devem ser realizados para validar o efeito da incorporação de solventes em infiltrantes experimentais no ângulo de contato com a estrutura dental (esmalte), uma vez que este estudo foi realizado em lâmina de vidro, o que variou o ângulo de contato em função dos monômeros utilizados. Embora alguns testes padrões, tenham sugerido mudanças nas propriedades, a incorporação de solventes nos infiltrantes é uma alternativa para melhorar significativamente a performance destes materiais. Estudos sobre a estabilidade de infiltrantes com adição de DMSO ou THF poderiam esclarecer o papel destes componentes na longevidade da infiltração na lesão de cárie.

## CONCLUSÃO

Diante dos resultados encontrados neste estudo, pode ser concluído que:

- 1- Icon<sup>®</sup> apresentou os melhores resultados dentre os materiais avaliados, com menor ângulo de contato, maior dureza e boa penetração.
- 2- Dentre os materiais experimentais, a incorporação de menores porcentagens dos solventes (0,5%) em infiltrantes com presença do UDMA apresentou resultados promissores nas propriedades físico-química dos infiltrantes experimentais, exceto no ângulo de contato.
- 3- O ângulo de contato dos infiltrantes experimentais variou em função da viscosidade dos monômeros.
- 4- A menor porcentagem de ambos os solventes associados a mistura 75% T + 25% UDMA, juntamente com o Icon, apresentaram os maiores valores de resistência de união.

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<sup>1</sup>*De acordo com a norma da UNICAMP/FOP, baseado na norma do International Committee of Medical Journal Editors – Grupo de Vancouver, Abreviatura dos periódicos em conformidade com o Medline,*



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## APÊNDICE

### Capítulo 1

O ângulo de contato dos materiais foi testado utilizando um viscosímetro, (figura1) ligado a um computador. As gotas dos materiais foram dispensadas em lâminas de vidro, por meio de micro seringa acoplada ao dispositivo, e dessa forma foram captadas imagens (figuras 2 e 3) através de uma câmera e transferidas para o software que fez as medidas das gotas e os valores do ângulo de contato.

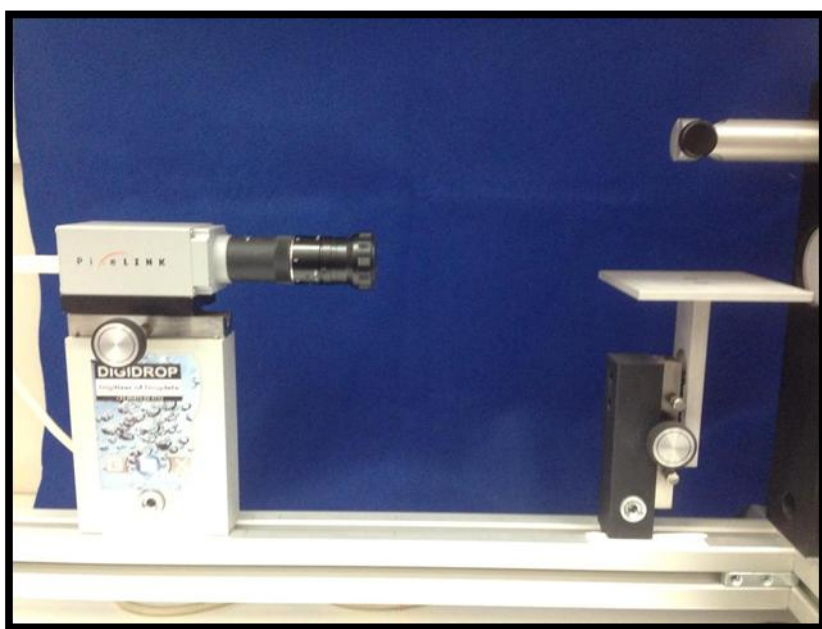


Figura 1. Viscosímetro (GBX Instrumentation Scientifique -France No, 04230711- Contact Angle Meter - Digitrop)

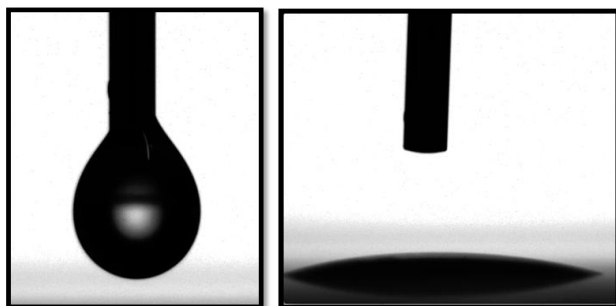


Figura 2. Imagem representativa do gota do grupo Icon<sup>®</sup>, obtida pela câmera para mensuração do ângulo de contato através do software.

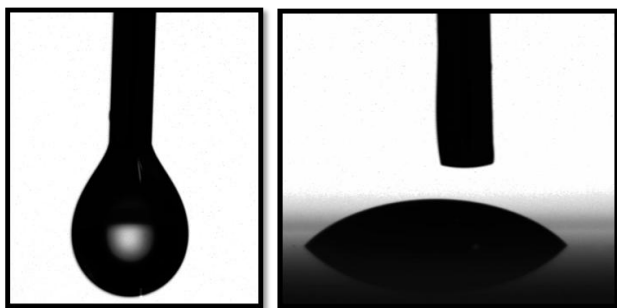


Figura 3. Imagem representativa do gota do grupo T + B + 0,5% DMSO, obtida pela câmera para mensuração do ângulo de contato através do software.

## Capítulo 2

Para o teste de resistência de união foram utilizados 10 dentes bovinos em cada grupo. Esses dentes após preparados foram infiltrados pelos materiais, com auxílio de uma matriz de silicone, formando blocos de material infiltrante sobre o esmalte artificialmente cariado de aproximadamente 5x5mm. Os conjuntos (dente-infiltrante) foram seccionados e fatias foram obtidas. As fatias foram preparadas em alta rotação com broca sob refrigeração e aproximadamente 4 halteres de cada dente eram confeccionados e levados a dispositivos específicos e acoplados na máquina de ensaio universal para mensuração da resistência de união (Figura 4). E posterior análise do padrão de fratura ( figuras 5, 6 e 7) em MEV.

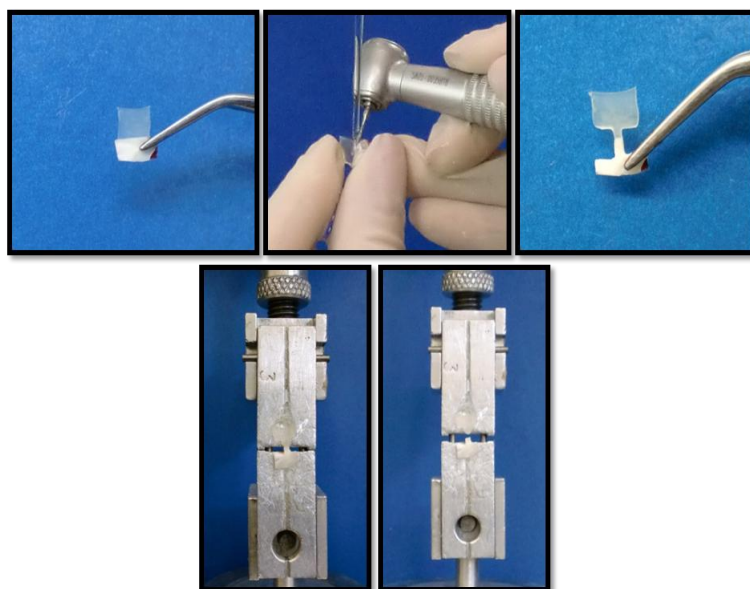


Figura 4. Imagem ilustrativa do preparo das amostras para mensuração da resistência de união

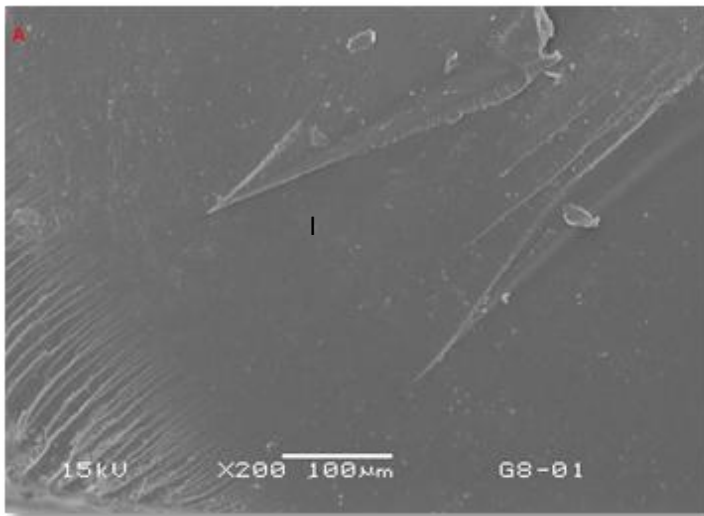


Figura 5: Imagem ilustrativa de fratura coesiva no infiltrante. I- Infiltrante.

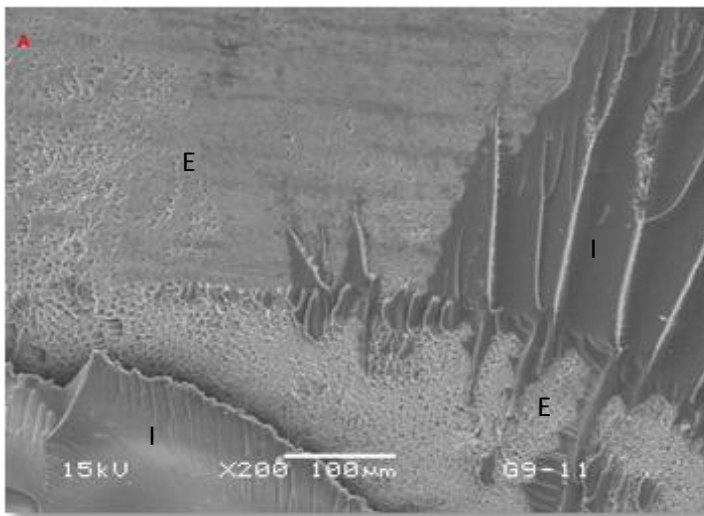


Figura 6. Imagem ilustrativa de fratura mista entre a camada de esmalte e infiltrante. E- Esmalte, I- Infiltrante.

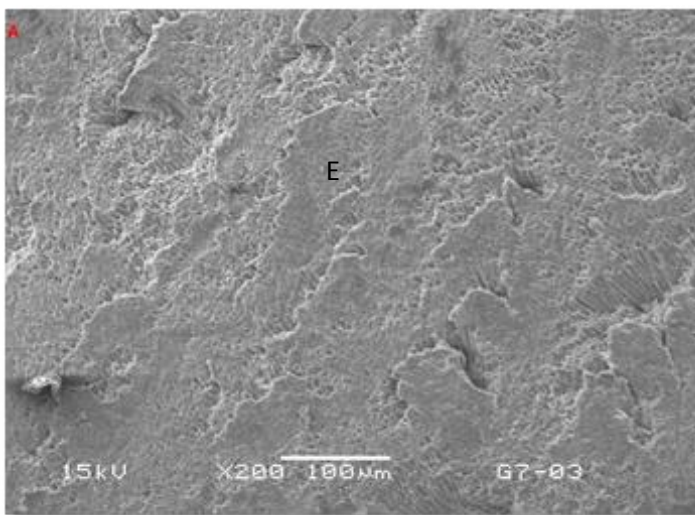


Figura 7. Imagem ilustrativa de fratura coesiva no esmalte. E- Esmalte.

Na mensuração do grau de conversão foi utilizado o aparelho FTIR (Nicolet 5700, Thermo Fisher Scientific, Loughborough, UK), equipado com um cristal de ATR. O material colocado sobre a base de FTIR ( $n = 3$ ) e alturas de picos foram registrados de acordo com os picos alifáticos e aromáticos (Figura 8a,b).

Após a leitura ( $n = 3$ ) a gota do material foi fotopolimerizada (60 segundos;  $1.000 \text{ mW/cm}^2$ , Optilux VLC, Demetron Kerr, Orange, EUA) e novas alturas de picos foram registrados (8c).

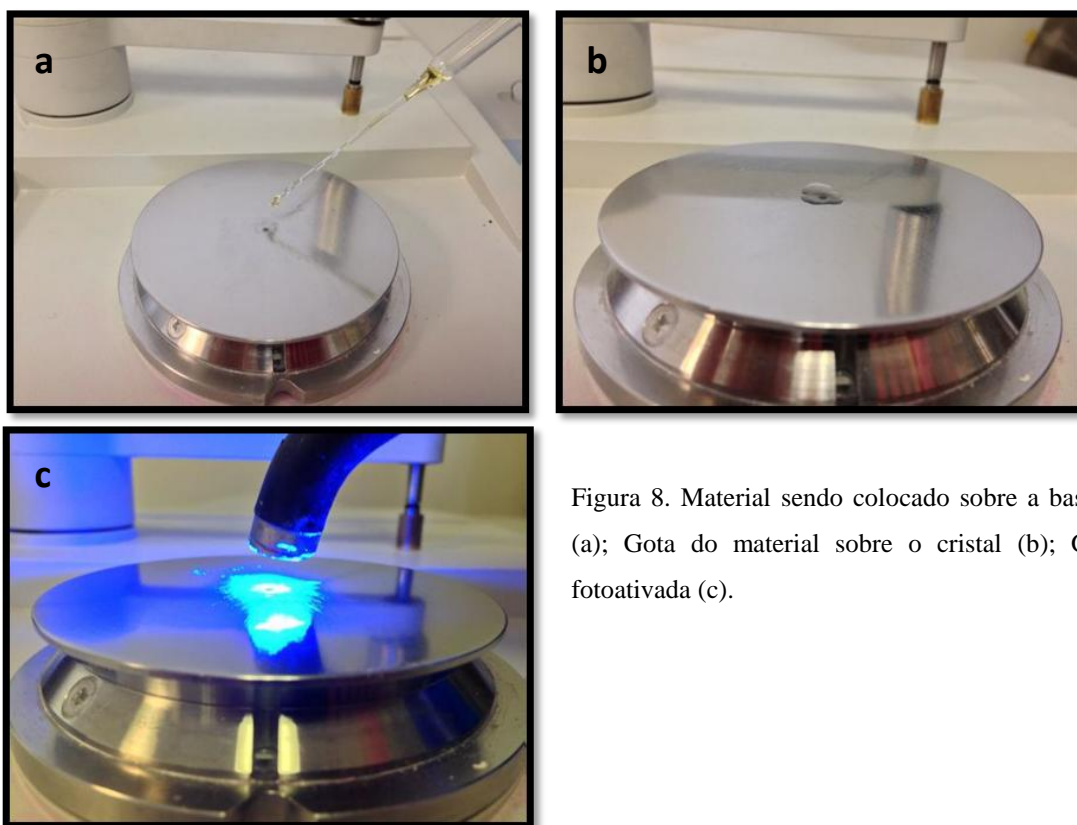


Figura 8. Material sendo colocado sobre a base do FTIR (a); Gota do material sobre o cristal (b); Gota sendo fotoativada (c).

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