UNIVERSIDADE ESTADUAL DE CAMPINAS
FACULDADE DE ODONTOLOGIA DE PIRACICABA

LUCIANA TIEMI INAGAKI

AVALIAÇÃO DE PROPRIEDADES FÍSICO-QUÍMICAS DE INFILTRANTES EXPERIMENTAIS COM ADIÇÃO DE CLOREXIDINA

EVALUATION OF PHYSICAL-CHEMICAL PROPERTIES OF EXPERIMENTAL INFILTRANTS CHLORHEXIDINE-ADDED

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Tese apresentada à Faculdade de Odontologia de Piracicaba da Universidade Estadual de Campinas como parte dos requisitos exigidos para obtenção do título de Doutora em Odontologia, na Àrea de Odontopediatria.

Thesis presented to the Piracicaba Dental School of the University of Campinas in partial fulfillment to the requirements for the degree of Doctor, in the area of Pediatric Dentistry.

Orientadora: Profa. Dra. Fernanda Miori Pascon

Coorientadora: Profa. Dra. Regina Maria Puppin-Rontani

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A Ata da defesa com as respectivas assinaturas dos membros encontra-se no processo de vida acadêmica do aluno.

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Cora Coralina

RESUMO

Este estudo objetivou avaliar os efeitos da clorexidina (CHX) e base monomérica nas propriedades físico-químicas de infiltrantes experimentais. Esta tese foi dividida em dois capítulos. Misturas monoméricas com duas concentrações de CHX foram preparadas: TEGDMA, TEGDMA/0,1%CHX, TEGDMA/0,2%CHX, TEGDMA/UDMA, TEGDMA/UDMA/0,1%CHX, TEGDMA/UDMA/0,2%CHX, TEGDMA/BisEMA, TEGDMA/BisEMA/0,1%CHX e TEGDMA/BisEMA/0,2%CHX. O infiltrante Icon® foi utilizado como controle comercial. O **Capítulo 1** avaliou a sorção/solubilidade (SS), taxa de redução de dureza (RD), módulo de elasticidade (EM) e resistência à flexão (FS) das nove misturas experimentais. A SS foi realizada conforme a ISO 4049, com discos de 7 mm de diâmetro x 1 mm de espessura ($n=5$); para RD utilizou-se discos com 5 mm de diâmetro x 1 mm de espessura ($n=10$), para EM e FS utilizou-se barras com 7 mm x 2 mm x 1 mm ($n=10$). Os dados foram submetidos à ANOVA e testes de Tukey ($\alpha=5\%$). A presença de CHX, independente da concentração, não interferiu nos resultados da SS, EM e FS. Para sorção, as misturas à base de TEGDMA apresentaram médias maiores e foram diferentes do controle ($p<0,01$). Misturas à base de TEGDMA/BisEMA apresentaram médias menores e não diferiram do controle ($p>0,05$). Misturas à base de TEGDMA/UDMA mostraram valores intermediários e diferiram do controle ($p<0,01$). Quanto à solubilidade, Icon® foi o mais solúvel comparado às misturas experimentais ($p<0,01$). Misturas com UDMA e BisEMA foram menos solúveis e não diferiram estatisticamente entre si ($p>0,05$). Quanto ao EM, todas as misturas diferiram do controle ($p<0,01$), que mostrou menor média. Misturas com UDMA apresentaram as maiores médias, não diferindo entre si e de TEGDMA/BisEMA ($p>0,05$). Para FS as misturas com UDMA e BisEMA foram diferentes do controle ($p<0,01$), apresentando as maiores médias. Misturas à base de TEGDMA e Icon® não foram diferentes entre si ($p>0,05$), com os menores valores de FS. Para RD, a mistura TEGDMA/UDMA/0,1%CHX apresentou a maior média, diferindo dos materiais experimentais ($p<0,05$) e não diferiu do controle ($p>0,05$). O **Capítulo 2** avaliou, por meio de perfilometria sem contato de superfície, a rugosidade de superfície em 2D (Ra) e em 3D (Sa) e a perda de material após desgaste mecânico por três corpos ($n=8$). Para isso, foram avaliadas as misturas: TEGDMA/0,2%CHX; TEGDMA/UDMA/0,2%CHX, TEGDMA/BisEMA/0,2%CHX e o controle comercial (Icon®). Os dados foram submetidos à ANOVA para medidas repetidas, ANOVA um fator, teste de Tukey e regressão linear simples (α=5%). Após desgaste mecânico, a mistura TEGDMA/UDMA/0,2%CHX mostrou menores valores de rugosidade (Ra e Sa) e menor perda de estrutura diferindo do controle ($p<0,01$). A mistura TEGDMA/BisEMA/0,2%CHX mostrou maior desgaste diferindo do controle ($p<0,01$). Houve associação significativa ($p<0,01$) entre rugosidade em Ra e em Sa (R^2=74,56%). Diante dos resultados concluiu-se que a adição de CHX não afetou negativamente as propriedades de SS, EM e FS. A presença de 0,1% de CHX afetou a RD da mistura com UDMA. A base monomérica das misturas influenciou as propriedades avaliadas nos Capítulos 1 e 2. A mistura à base de TEGDMA/UDMA mostrou os melhores resultados para as propriedades avaliadas, exceto para RD. Medidas da rugosidade em Ra e Sa mostraram associação.

Palavras-chave: Clorexidina. Escovação dentária. Módulo de elasticidade. Materiais dentários. Solubilidade.

ABSTRACT

This study evaluated the chlorhexidine (CHX) and monomeric base effects in physical-chemical properties of experimental resin materials. This thesis was divided in two chapters. Monomeric blends with two concentrations of CHX were prepared: TEGDMA, TEGDMA/0.1%CHX, TEGDMA/0.2%CHX, TEGDMA/UDMA, TEGDMA/UDMA/0.1%CHX, TEGDMA/UDMA/0.2%CHX, TEGDMA/BisEMA, TEGDMA/BisEMA/0.1%CHX e TEGDMA/BisEMA/0.2%CHX. Icon® infiltrant was used as commercial control group. The **Chapter 1** evaluated sorption/solubility (SS), softening, elastic modulus (EM) and flexural strength (FS) of nine experimental blends. The SS was accomplished according to ISO 4049, using disks with 7 mm of diameter x 1 mm of thick (n=5); for softening, disks with 5 mm of diameter x 1 mm of thick were used (n=10); for EM and FS bars with 7 mm x 2 mm x 1 mm were used (n=10). The data were submitted to ANOVA and Tukey test ($\alpha=5\%$). The CHX presence, regardless concentration, did not interfere in SS, EM and FS. For sorption, the TEGDMA blends showed the highest averages and they were different than control ($p<0.01$). The TEGDMA/BisEMA blends presented the lowest averages and they were not different than control ($p>0.05$). TEGDMA/UDMA based blends showed intermediate values and they were different than control ($p<0.01$). Concerning solubility, Icon® was the most soluble when it was compared with experimental blends ($p<0.01$). UDMA and BisEMA based blends were less soluble and they did not differ statistically between each other ($p>0.05$). About EM, all blends were different than control ($p<0.01$), which show the lowest average. UDMA blends showed the highest average, and they did not differ between each other and from TEGDMA/BisEMA ($p>0.05$). For FS, the blends with UDMA and BisEMA were different than control ($p<0.01$), showing the highest average. TEGDMA blends and Icon® were not different between each other ($p>0.05$), with the lowest values of FS. For softening, TEGDMA/UDMA/0.1%CHX showed the highest average and it was different than experimental blends ($p<0.05$), but it was not different than control ($p>0.05$). The **Chapter 2** evaluated using non-contact profilometer the surface roughness in 2D (Ra) and in 3D (Sa), and the material loss after mechanical wear by three-body (n=8). For this, the blends evaluated were: TEGDMA/0.2%CHX; TEGDMA/UDMA/0.2%CHX; TEGDMA/BISEMA/0.2%CHX and the commercial control (Icon®). The data were submitted to ANOVA for repeated measures, one-way ANOVA and Tukey and simple linear regression ($\alpha=5\%$). After mechanical wear, the TEGDMA/UDMA/0.2%CHX blend showed the lowest values for roughness (Ra and Sa) and for material loss, and it was different than control ($p<0.01$). The blend TEGDMA/BisEMA/0.2%CHX showed the highest mechanical wear and it was different than control ($p<0.01$). There was significant association ($p<0.01$) between Ra and Sa roughness ($R^2=74.56\%$). In front of the results it could be concluded that the CHX did not affect negatively the SS, EM and FS properties. The presence of 0.1% of CHX affected UDMA blend softening. The blends monomeric base influenced the properties evaluated in Chapter 1 and 2. The TEGDMA/UDMA based blends showed the better results for properties evaluated, except for softening. Superficial roughness in Ra and Sa showed association.

KEY WORDS: Chlorhexidine. Toothbrushing. Elastic modulus. Dental materials. Solubility.

SUMÁRIO

1. INTRODUÇÃO

A doença cárie dentária tem apresentado declínio significante na população de países desenvolvidos e em desenvolvimento, mas continua sendo um problema de saúde entre pré-escolares e adolescentes (James et al., 2010; Chankanka et al., 2011; Freitas et al., 2014). Maus hábitos alimentares, principalmente relacionados ao consumo frequente e excessivo de sacarose, associados à remoção inadequada do biofilme presente na superfície dentária, fazem com que a doença cárie seja um dos maiores problemas epidemiológicos do mundo (Sheiham e James, 2015). Indivíduos jovens com dentes recém erupcionados, principalmente na fase de dentição mista, estão mais predispostos ao desenvolvimento de lesões cariosas (James et al., 2010; Chankanka et al., 2011; Fontana et al. 2011; Sarmadi et al., 2011).

A lesão de cárie se inicia a partir de um desequilíbrio iônico entre o esmalte e a saliva produzido pelo biofilme cariogênico aderido à superfície do esmalte (Gray e Shellis, 2002). Os estágios iniciais da dissolução do esmalte envolvem desintegração da superfície, com espaços intercristalinos mais ampliados levando, até mesmo, a formação de microcavidades (Gray e Shellis, 2002). As lesões de cárie se desenvolvem em locais propícios para depósitos bacterianos como regiões interproximais, margem gengival e superfícies oclusais de dentes posteriores (Robinson et al., 2001; Gray e Shellis, 2002). Durante o desenvolvimento da lesão cariosa, o mineral é removido da estrutura do esmalte deixando-o com porosidades, visualmente reconhecidas como manchas brancas opacas (Robinson et al., 2001; Gray e Shellis, 2002).

Na lesão de mancha branca, a zona superficial do esmalte aparece relativamente intacta, com cerca de 8% de perda mineral, enquanto nas camadas subjacentes a perda pode variar de 20 a 90% e o volume dos poros se encontra aumentado (Bergman e Lind, 1966). Essa hipermineralização da camada superficial da lesão de mancha branca torna esse esmalte mais resistente ao condicionamento ácido (Lee et al., 1995). O método mais amplamente utilizado e comprovadamente eficaz para cessar a progressão da lesão inicial de cárie em esmalte tem sido o uso de fluoretos em suas mais variadas formulações e concentrações (Tenuta e Cury, 2010). Apesar da eficiência na remineralização do esmalte desmineralizado, a colaboração do paciente tem papel fundamental para a atuação dos fluoretos, uma vez que seu uso está associado às boas práticas de higiene bucal, o que muitas vezes não é possível (Leal, 2014).

Atualmente, os conceitos de mínima intervenção para os tratamentos restauradores tem ganhado espaço na Odontologia, pois há o intuito de preservar ao máximo a estrutura dental (Leal, 2014; Freitas et al., 2014). Assim, a paralisação da progressão da cárie incipiente utilizando materiais infiltrantes, como monômeros resinosos fotoativáveis altamente fluidos, é uma alternativa para a prática da Odontologia minimamente invasiva. O conceito de infiltração das lesões iniciais de cárie não é novo, e estudos pioneiros avaliaram a capacidade de infiltração em lesões naturais e artificiais de esmalte por meio de adesivo (Dávila et al., 1975) e formulações de resorcinol-formaldeído (Robinson et al., 1976). A partir do princípio do selamento oclusal, pensou-se numa alternativa para o tratamento das lesões iniciais não cavitadas em esmalte utilizando sistema adesivo (Garcia-Godoy et al., 1997), e comparando um sistema adesivo com selante resinoso, sendo ambos materiais considerados capazes de penetrar os poros do esmalte desmineralizado (Gray e Shellis, 2002).

Paris et al. (2007a) analisaram várias combinações de monômeros (hidrófobos e hidrófilos) e solventes com a finalidade de verificar o coeficiente de penetração das misturas. Com a adição de etanol, os autores notaram que o coeficiente de penetração das misturas aumentou significativamente devido à diminuição da viscosidade e do ângulo de contato, e os maiores coeficientes de penetração foram obtidos para as misturas que continham trietilenoglicol dimetacrilato (TEGDMA), metacrilato de hidróxi etila (HEMA) e 20% de etanol. Assim, o uso de um material resinoso altamente fluido (infiltrante) para a penetração dos poros do corpo da lesão de mancha branca é fator imprescindível para que a capacidade de inibir a desmineralização do esmalte seja efetiva; e a presença do material resinoso preenchendo os poros ampliados da lesão pode estabilizar a estrutura frágil do esmalte comprometido por meio de um reforço mecânico estrutural (Paris et al., 2007a).

Para melhor penetração dos infiltrantes nas lesões cariosas naturais não cavitadas, há a necessidade de condicionamento ácido prévio para a erosão da camada superficial hipermineralizada que é também contaminada por água e substâncias orgânicas (Meyer-Lueckel, et al., 2007; Meyer-Lueckel e Paris, 2008; Paris e Meyer-Lueckel, 2010). Dessa forma, a aplicação de ácido clorídrico a 15% por 120 segundos apresentou melhores resultados quando comparado ao uso do ácido fosfórico a 37% na penetração de adesivo no esmalte dental (Paris *et al.*,2007b). Entretanto, resultados satisfatórios quanto à penetração de materiais infiltrantes experimentais foram observados quando diferentes tempos (15, 30, 60 e 120 segundos) de condicionamento ácido com ácido fosfórico a 35% foram utilizados em lesões artificiais de cárie em esmalte bovino. Neste estudo, o condicionamento ácido por 15 segundos proporcionou condições ideais para a penetração de infiltrantes experimentais (Araújo, 2011a).

Materiais com alto coeficiente de penetração, denominados de "infiltrantes" passaram a ser caracterizados até a formulação do Infiltrante Icon® (DMG, Hamburgo, Alemanha), disponível comercialmente. Estudos foram conduzidos com o objetivo de avaliar a afetividade deste material e foram mostrados resultados parcialmente satisfatórios, pois quando o infiltrante foi comparado clinicamente com o sistema adesivo nanoparticulado Prime & Bond NT (Dentstply, York, PA, USA) em lesões proximais (Martignon et al., 2012) ou com o selante de fóssulas e fissuras Delton (Dentsply Detrey, Copenhagen, Dinamarca), em lesões oclusais (Bakhshendeh e Ekstrand, 2015), não mostrou diferenças estatisticamente significativas. Resultados quanto à efetividade do Icon® em relação à estética (Borges et al., 2014; Araújo et al., 2015), lisura de superfície (Mueller et al., 2011; Belli et al., 2011) e adesão de biofilme (Arslan et al., 2015) também foram elucidados, o que instigou a necessidade de opções no mercado para novas formulações de materiais com capacidade infiltrante.

Misturas resinosas experimentais com características infiltrantes à base de TEGDMA, uretano dimetacrilato (UDMA) e bisfenol A etoxilado dimetacrilato (BisEMA) com a adição de diluentes como o etanol e HEMA, apresentaram redução da viscosidade, no entanto esses diluentes reduziram as propriedades físico-mecânicas como grau de conversão, dureza Knoop e o módulo de elasticidade (Araújo et al., 2013). Esses mesmos autores mostraram que as misturas sem adição de solventes apresentaram os melhores resultados para o grau de penetração, densidade de ligações cruzadas e resistência de união, sendo consideradas misturas promissoras para compor um material com características infiltrantes (Araújo et al., 2013; Araújo et al., 2015).

Para que materiais resinosos infiltrantes sejam realmente efetivos e apresentem excelência clínica, algumas características deveriam estar presentes como: a) ser um material hidrofílico; b) ser fluido e ter alto grau de penetração; c) ter ação antimicrobiana; d) não ser tóxico aos tecidos bucais; e) ser polimerizável; f) ser resistente aos desgastes químicos e mecânicos dentro da cavidade bucal; g) ser estético (Kielbassa et al., 2011). Dentre as várias funções necessárias para um material infiltrante, a adição de um agente antibacteriano às formulações experimentais à base de TEGDMA, UDMA e BisEMA poderia ser avaliada, uma vez que a presença de um antimicrobiano na composição de materiais restauradores poderia diminuir ou impedir a adesão de biofilme na superfície da restauração (Imazato, 2003).

A adição do diacetato de clorexidina (CHX) como agente antibacteriano às misturas resinosas com características infiltrantes à base de TEGDMA, UDMA e BisEMA foi proposto por Inagaki (2012). A adição de CHX em concentrações de 0,1% e 0,2% mostrou atividade antibacteriana contra cepas de *Streptococcus mutans* e *Lactobacillus acidophillus* e não afetou negativamente propriedades como grau de conversão e dureza Knoop (Inagaki, 2012).

A clorexidina é considerada uma molécula catiônica simétrica por possuir dois anéis 4-clorofenis e dois grupos biguanidas ligados à cadeia central de hexametileno. Por ser uma base forte, a molécula da clorexidina é mais estável na forma de sal. Devido as suas propriedades catiônicas, a clorexidina se liga à hidroxiapatitia do esmalte dentário, a película adquirida na superfície dentária, as proteínas salivares, as bactérias e as proteínas extracelulares de origem bacteriana, apresentando um amplo espectro de ação contra fungos, cepas gram-positivas e gram-negativas (Fardal e Turnbull, 1986). Entre as bactérias gram-positivas, os *Streptococcus mutans* são particularmente mais sensíveis à clorexidina que as espécies *Lactobacillus sp* (Emilson, 1994).

O mecanismo de ação da clorexidina reside na capacidade de se adsorver na parede celular do microrganismo que provoca a liberação de componentes intracelulares (Fardal e Turnbull, 1986). Em baixas concentrações, a clorexidina provoca a liberação de substâncias com baixo peso molecular, como potássio e fósforo, exercendo um efeito bacteriostático (Fardal e Turnbull, 1986). Por outro lado, em altas concentrações, a clorexidina possui efeito bactericida devido à precipitação e coagulação do citoplasma, provavelmente causado por ligações cruzadas protéicas (Fardal e Turnbull, 1986). Deste modo, a clorexidina pode interferir na formação do biofilme dentário: por desativar a enzima glicosiltransferase secretada pelo *Streptococcus mutans*, enzima importante na aderência bacteriana à superfície dentária; e também por deslocar cálcio dos grupos sulfatos, desintegrando o biofilme já estabelecido (Ribeiro et al., 2008). Assim, devido ao seu amplo espectro de ação e capacidade de inibir a formação do biofilme, a clorexidina tem sido utilizada no tratamento e prevenção de doenças periodontais e cárie dentária (Autio-Gold, 2008; Van Rijkom et al., 1996; Emilson, 1994).

Estudos têm sido realizados com sais de CHX (gluconato de clorexidina e diacetato de clorexidina) adicionados aos cimentos de ionômero de vidro convencionais, aos cimentos de ionômero de vidro modificados e aos materiais resinosos com o intuito de aumentar a efetividade clínica por meio da atividade antibacteriana (Riggs et al., 2000; Cacciafesta et al., 2006; Mehdawi et al., 2009; Hiraishi et al., 2010; Tüzüner et al., 2011). A incorporação de CHX aos sistemas adesivos utilizados na colagem de braquetes mostrou bons resultados quanto à resistência ao cisalhamento (Ribeiro et al., 2008, Bishara et al., 1998; Damon et al., 1997). Assim, a incorporação de CHX como agente antimicrobiano na composição de materiais infiltrantes experimentais é uma proposta promissora, pois poderia aumentar a efetividade desses materiais quanto à atividade antimicrobiana, principalmente em relação aos microrganismos cariogênicos residuais presentes nas lesões cariosas incipientes. Além disso, essa nova proposta de materiais infiltrantes poderia diminuir a colonização bacteriana do biofilme sobre a área infiltrada, protegendo o esmalte sadio adjacente.

Apesar do propósito dos infiltrantes não ser a restauração de lesões cariosas e sim, promover a paralisação de lesões iniciais não cavitadas preenchendo os poros de esmalte, testes de resistência à degradação mecânica devido ao desgaste pelo processo de escovação são importantes, uma vez que esses materiais estarão em contato direto com a região superficial do esmalte (faces interproximais, vestibulares e oclusais) e expostos à desafios químicos e físicos da cavidade bucal.

O aumento da rugosidade na superfície de materiais restaurados propicia um maior acúmulo de biofilme dental; causando biodegradação superficial e, consequentemente, comprometendo a vida útil da restauração (Santos et al., 2007; De Paula et al., 2011).

A interação de materiais resinosos com as condições úmidas da cavidade bucal provoca redução de suas propriedades mecânicas, pois os compostos poliméricos utilizados nas superfícies dentais tendem a absorver água (mecanismo de sorção que causa um aumento de massa) e liberar monômeros não polimerizados por meio da lixiviação de partículas residuais que leva a uma redução de massa (Sideridou et al., 2011). A capacidade de um polímero sorver e liberar componentes para o meio líquido pode estar relacionado com as características químicas dos monômeros, com a interação molecular após polimerização, bem como com as propriedades elásticas do polímero para expandir pela captação de um solvente (Sideridou et al., 2003; Ferracane, 2006).

O monômero TEGDMA tem sido identificado como a principal substância liberada pelas resinas compostas, porém, pequenas quantidades de outros monômeros, tais como bisfenol A glicidil metacrilato (BisGMA) e UDMA podem também ser liberados na água, comprometendo a durabilidade do material restaurador (Örtengren *et al.*, 2001). A CHX incorporada em materiais resinosos e em cimentos ionoméricos também pode ser liberada para o meio bucal e isso pode influenciar diretamente na ação antimicrobiana e interferir nas propriedades físicas e mecânicas do material (Leung et al., 2005; Anusavice et al., 2006; Hiraishi et al., 2008).

Dessa forma, diante do exposto acima, esta tese objetivou: 1) avaliar as propriedades físico-químicas, como sorção/solubilidade, taxa de redução de dureza e módulo de elasticidade/resistência à flexão de infiltrantes experimentais com diferentes monômeros e adição de CHX; e 2) avaliar a resistência de infiltrantes experimentais, com diferentes monômeros e adição de CHX, ao desgaste mecânico por três corpos. Para atingir esses objetivos, esta tese foi dividida em dois capítulos, os quais serão apresentados a seguir.[1]

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[1] Esta tese foi redigida no formato alternativo, com base na resolução da CCPG/001/2015, a qual dispõe a respeito do formato das dissertações de mestrado e teses de doutorado aprovados pela UNICAMP.

2. ARTIGOS

2.1 ARTIGO 1

Evaluation of sorption/solubility, softening, flexural strength and elastic modulus of experimental resin blends with chlorhexidine[2]

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ABSTRACT

Objectives: To evaluate physical-chemical properties of experimental diacetate chlorhexidine (CHX)-added resin blends.

Methods: Blends were formulated: G1)TEGDMA; G2)TEGDMA/0.1%CHX; G3)TEGDMA/0.2%CHX; G4)TEGDMA/UDMA; G5)TEGDMA/UDMA/0.1%CHX; G6)TEGDMA/UDMA/0.2%CHX; G7)TEGDMA/BisEMA, G8)TEGDMA/BisEMA/0.1%CHX; G9)TEGDMA/BisEMA/0.2%CHX. Icon® was the control group. For sorption/solubility (SS), cylindrical specimens (n=5) were prepared and their weight obtained. The specimens were immersed in deionized water for 7 days at 37°C and their weight was verified again. SS were calculated using accepted formulas. For softening, cylindrical specimens (n=10) were prepared and initial Knoop hardness number (KHN) obtained. The specimens were immersed in absolute ethanol for 24 h at 37°C and final KHN accomplished. Softening values were calculated by KHN reduction percentage. For elastic modulus (EM) and flexural strength (FS) bar specimens were prepared (n=10) and values obtained with a universal device (three point, 5 mm distance, 0.5 mm/min, load of 50 N). The data was analyzed using one-way ANOVA and Tukey test (α=5%).

Results: TEGDMA/BisEMA blends and Icon® showed the lowest sorption from blends ($p>0.05$), and Icon® was the most soluble material ($p<0.01$). TEGDMA/UDMA/0.1%CHX showed the highest softening, similar to Icon® ($p>0.05$). For EM, all blends were different than Icon® ($p<0.01$). For FS, TEGDMA blends were similar to Icon®, showing the lowest averages ($p>0.05$).

Conclusions: Monomers chemical characteristics influenced the physical-chemical properties of experimental blends more than CHX. Between the blends tested, UDMA blends presented satisfactory results for assays evaluated.

Clinical Significance: Infiltrants CHX-added could arrest and reinforce initial caries lesions, and the antimicrobial effect could prevent new lesions in sound enamel adjacent to the infiltrated area.

KEY WORDS: Sorption, solubility, softening, flexural strength, elastic modulus, chlorhexidine, infiltrants.

1. INTRODUCTION

The main objective of minimal invasive dentistry is to preserve the dental structure with less invasive caries management strategies.[1] Previously studies showed the possibility to infiltrate initial caries lesions with low viscosity materials.[2,3] The principle of sealing white spot lesions with an unfilled resin was reported as a potential effectiveness method for minimal intervention.[4] Therefore, the infiltrants are a class of dental materials developed to arrest the progression of initial caries lesion and to reinforce mechanically the fragile demineralized enamel.[5,6] Actually, a TEGDMA (triethylene glycol dimethacrylate) based-material (Icon®, DMG, Hamburg, Germany) is a resin material available commercially with infiltrant functions. This material was developed from studies with experimental resin blends containing different concentrations of monomers as TEGDMA and BisGMA (bisphenol A diglycidyl methacrylate) with different concentrations of ethanol, and blends containing high amount of TEGDMA with ethanol had high coefficient degrees.[5] Nevertheless, high coefficient of penetration in natural lesions was obtained with solvent-free TEGDMA blends.[7]

The Icon® effectiveness to penetrate initial caries lesions was evaluated in *in vitro* studies[8,9] and clinical trials studies reported it capacity to arrest lesion progression.[10,11] Studies showed satisfactory esthetic results with Icon® masking white spots in enamel,[12,13] the infiltrant also have been studied for controlling enamel erosion progression.[14] Nevertheless, a clinical trial study with 3 years of follow up, comparing the effectiveness of proximal caries infiltration with Icon® and with a nanoparticulate fluoride-releasing adhesive system, showed that both materials had similar performance and the infiltration technique was better than the control group, which received only oral health instructions.[15]

Recently, experimental low viscosity monomer blends with infiltrants characteristics containing TEGDMA, UDMA (urethane dimethacrylate) and BisEMA (bisphenol A ethoxylate dimethacrylate) using diluents as ethanol or HEMA (2-hydroxyethyl methacrylate) have been tested in *in vitro* studies.[16] Concerning physical and mechanical properties, such as degree of conversion, elastic modulus, Knoop hardness and softening ratio, these experimental infiltrants were similar or better when compared to an infiltrant commercially available.[16] These formulations proposed for infiltrant materials seem promising and the addition of an antimicrobial agent could improve their clinical performance decreasing biofilm growth over surfaces and, consequently, avoiding harmful deteriorations.

The advantages of adding antimicrobial agents into resin materials, as composites and dentin bonding systems, to avoid biofilm formation around the restoration had been already discussed in a review,[17] and the use of chlorhexidine (CHX) could be an option. The addition of CHX in dental resin materials was reported in studies that showed the possibility to incorporate 1% CHX into resin materials without considerable changes in their physical-mechanical properties, such as degree of conversion, elastic modulus[18] and bond strength.[19] However, in a humid environment, when the CHX is entrapped in the resin bulk after polymerization, some of the antimicrobial and unpolymerized particles could extrude from the polymer matrix, which could influence the mechanical and physical properties of the material.[20,21]

The mechanical properties of resin materials could be altered by hydrolysis, and the quality of polymerization could be related to the chemical characteristics of the monomers. The resin materials properties are influenced by water present in oral environment.[22] The monomers as UDMA, TEGDMA and BisEMA have carbon and oxygen or nitrogen in their structures, with hydrolytic susceptible groups, such as ester, urethane, ether linkages, as well as hydroxyl groups.[23] The phenomenon of sorption and solubility may serve as precursors to a variety of chemical and physical processes that include volumetric (such as swelling), physical (such as plasticization), softening and chemical (such as oxidation and hydrolysis) changes.[23] The experimental materials with infiltrants characteristics, as well as other restorative resin dental materials, could be exposed to a wet environment suffering changes. Sorption and solubility properties are important to evaluate the hydrolytic degradation of resin materials. The resistance of water degradation could be associated to the chemical characteristics of the polymers, including quality of cross-link density into bulk matrix and softening ratio.[24-27]

Therefore, the aim of this study was to evaluate the physical-chemical properties (sorption/solubility, softening and elastic properties) of experimental resin blends with CHX. The first hypothesis tested was that physical-chemical properties could be influenced by different composition of experimental blends and the second hypothesis was that different concentration of CHX would not interfere in the properties evaluated.

2. MATERIAL AND METHODS

2.1 Formulation of low viscosity monomers blends

Experimental resin blends were made using the composition shown in Tables 1 and 2. The monomer TEGDMA was used as a main component for all resin blends. The monomers UDMA or BisEMA were used in some blends in a proportion of 1:4 (wt/wt) (Table 1). The photoinitiator system used in the blends was 1.0 wt % of DMAEMA (2-dimethylaminoethyl methacrylate) and 0.5 wt % of CQ (camphorquinone). The inhibitor BHT (butylated hydroxytoluene) was used in a proportion of 0.1 wt %. Two different concentrations (0.1 wt % and 0.2 wt %) of CHX were tested (Table 1). All chemical components were weight individually at an analytical balance (Mark 210A, BEL Engineering, Piracicaba, SP, Brazil) and the blends were prepared at room temperature in a dark environment. Experimental infiltrants were mixed with a spatula in a beaker. In order to prevent premature polymerization, the resins were stored in the dark and opaque recipients, protected from light, at 4°C until use. The infiltrant Icon® (DMG – Hamburg, Germany, Batch 666352), was used as commercial control group.

Table 1. Composition of low viscosity monomer blends with infiltrant characteristics.

Experimental blends	Composition
G1	TEGDMA (100 wt.%)
G2	TEGDMA (99.9 wt.%), CHX 0.1 wt.%
G3	TEGDMA (99.8 wt.%), CHX 0.2 wt.%
G4	TEGDMA (75 wt. %), UDMA (25 wt. %)
G5	TEGDMA (74.9 wt.%), UDMA (25 wt.%), CHX 0.1 wt.%
G6	TEGDMA (74.8 wt.%), UDMA (25 wt.%), CHX 0.2 wt.%
G7	TEGDMA (75 wt.%), BisEMA (25 wt.%)
G8	TEGDMA (74.9 wt.%), BisEMA (25 wt.%), CHX 0.1 wt.%
G9	TEGDMA (74.8 wt.%), BisEMA (25 wt.%), CHX 0.2 wt.%
Icon®	TEGDMA, initiators and stabilizers

Table 2. Monomers and photoinitiators used for formulations of resin blends.

Chemical Component	CAS Number	Molecular Weight g/mol	Molecular Formula	Manufacturer	Batch
TEGDMA	109-16-0	286.32	$C_{14}H_{22}O_6$	Sigma-Aldrich, St. Louis, MO, USA	36296HK
UDMA	72869-86-4	470.56	$C_{23}H_{38}N_2O_8$	Sigma-Aldrich, St. Louis, MO, USA	MKBD1130
BisEMA	41637-38-1	376.4	$C_{21}H_{28}O_6$	Sigma-Aldrich, St. Louis, MO, USA	03514HF
CQ	10373-78-1	166.22	$C_{10}H_{14}O_2$	Sigma-Aldrich, St. Louis, MO, USA	083K0014
BHT	128-37-0	220.35	$C_{15}H_{24}O$	Sigma-Aldrich, St. Louis, MO, USA	04416KD
DMAEMA	2867-47-2	157.21	$C_8H_{15}NO_2$	Sigma-Aldrich, St. Louis, MO, USA	1437599
CHX	56-95-1	625.55	$C_{22}H_{30}Cl_2N_{10}.2C_2H_4O_2$	Sigma-Aldrich, St. Louis, MO, USA	083K0014

2.2 Sorption and Solubility Test

Sorption and solubility evaluation were performed according to ISO 4049,[28] except for specimens dimensions. Five resin disks (n=5) were made for each material using a polyvinyl siloxane mold (Express XT, 3M ESPE, St. Paul, MN, US) with 7 mm of diameter and 1 mm of thick. The diameter was used with intention to promote homogeneous polymerization, covering all specimen surfaces with active tip of photocuring device. The mold was completely filled with the blend. After this, a polyester strip was placed over and covered with a glass slide until light curing in order to obtain a smooth and standard flat surface. Each disk was light cured for 60 s with LED VALO device (Ultradent, South Jordan, UT, USA) with power density of approximately 1000 mW/cm^2. Immediately after polymerization, the disks were stored, individually, in closed Eppendorfs at 37°C in dry conditions for 24 h. After this period, the Eppendorfs containing the disks were opened and placed in a desiccator containing silica gel in a vacuum environment at 37°C for 22 h. After this period, the silica gel was changed and the set was kept at 37°C for more 2 h. When the 24 h of storage at desiccator were completed, each disk was weighed in an analytical balance

(Tel Marke, Bel Quimi, São Paulo, SP, Brazil) with an accuracy of 0.001 g. This cycle of drying in silica and weighing in a balance was repeated until a constant mass (M1) of each disk was obtained. For this, the mass of each disk should not have a variation greater than 0.001 g at interval of 24 h period. In this study, after 4 days of consequently weighing, the M1 of disks was established.

Immediately after M1 establishment, the measurements of two diameters of disks were taken using a digital caliper (0.01-150 mm, Product Code 500-144B, Mitutoyo, Tokyo, Japan) with an accuracy of 0.01mm. The same procedure was done to record thickness values. The mean value of diameter and thickness were obtained to calculate the volume of the cylinder (V), in cubic millimeters (mm^3). Then, the disks were immersed, individually, in falcon tubes with 4.66 mL of deionized water at 37°C for 7 days. The final water volume was calculated taking the proportion suggested by ISO: 10 mL of water for each disk with 15 mm of diameter and 1 mm of depth.

After 7 days, the disks were removed from the water and dried in absorbent paper for 15 s. One minute after removal from the water, each disk was weighed only one time in an analytical balance to obtain the M2 mass. After this, the disks were placed again in a vacuum desiccator with silica gel, and the same cycle already described was done until they kept a constant mass M3. For this study, 5 days of weighing were necessary to obtained the values of M3. The values (in $\mu g/mm^3$) of water sorption (W_{sp}) and solubility (W_{sl}) were calculated using the following equation: $W_{sp} = (M_2 - M_3) / V$ and $W_{sl} = (M_1 - M_3) / V$.

2.3 Softening Test

For this test, disks (n=10) with 5 mm diameter and 1 mm thick were made for each experimental resin blends and the commercial control group (Table 1). A cylindrical mold of polyvinyl siloxane (Express XT, 3M ESPE, St. Paul, MN, US) was used to make the disks and the procedure of polymerization followed the same requirements used with to sorption/solubility disks. After polymerization, the disks were kept in dry environment at 37°C for 24 h.

The initial Knoop hardness number (KHN1) was taken on superficial surface by five indentations for each disk. Measurements were obtained with the indenter

Future-Tech FM-100 (Future-Tech Corp., Kawasaki-City, Japan) automatic procedure with a load of 10 gF applied for 5 s, using 10X magnification lens. The average of five indentations was considered for statistical analysis. After KHN1, the disks were immersed, individually, in 1 mL of absolute ethanol for 24 h at 37°C. After this period, the final Knoop hardness number (KHN2) was measured following the same procedure previously described. The softening was determined (in percentage) following equation[16]: *Softening = 100 – [(KHN2 / KHN1) x 100].*

2.4 Flexural Strength and Elastic Modulus Tests

For these tests, bars specimens (7 mm x 2 mm x 1 mm) of each resin material (n=10) were prepared using a polyvinyl siloxane mold (Express XT, 3M ESPE, St. Paul, MN, US). The light curing process followed the same requirements already described. After this, the specimens were kept in a 100% humidity environment at 37°C for 24 h. The elastic modulus, in GPa, and flexural strength, in MPa, were performed using an universal testing device (Instron 4111, Instron Corp., Dayton, OH, USA) in a three point bending design with a crosshead speed of 0.5 mm/min and a cell load of 50 N until fracture. The distance between supports was 5 mm. The software Bluehill 2 (Illinois Tool Works, Inc., Glenview, IL, USA) was used to calculate the values of the tests.

2.5 Statistical Analysis

The data of sorption/solubility, softening and flexural strength/elastic modulus were analyzed with the Lilliefors test for the normality distribution. After this, one-way ANOVA and Tukey tests were applied to compare the groups at 5% level of significance. The statistical program used was Bioestat 5.3 (Mamirauá Institue, Tefé, AM, Brazil).

3. RESULTS

The results of SS, softening, EM and FS tests are shown in Table 3. For all tests, except for softening, the CHX concentration did not influence the experimental materials behavior. Concerning sorption test, TEGDMA/BisEMA resin blends groups (G7, G8 and G9) showed the lowest sorption values and they were not statistically different than the commercial infiltrant ($p>0.05$). TEGDMA resin blends (G1, G2 and G3) showed the highest values of sorption and they were not statistically different ($p>0.05$) between them and TEGDMA/UDMA blend (G4). The same behavior was noted for TEGDMA/UDMA blends (G4, G5 and G6) that showed intermediate values of sorption, but higher sorption than BisEMA blends and the commercial infiltrant ($p>0.05$).

Regarding the solubility test, all experimental blends (G1-G9) were statistically different than the commercial infiltrant ($p<0.01$), with low values of solubility. The commercial control group showed the highest value of solubility. TEGDMA blends (G1, G2 and G3) had higher solubility values and they were not statistically different ($p>0.05$) between them and the blends TEGDMA/UDMA/0.1CHX (G5), TEGDMA/BisEMA/0.1CHX (G8) and TEGDMA/BisEMA/0.2CHX (G9). TEGDMA/UDMA blends and TEGDMA/BisEMA blends did not show statistically differences ($p>0.05$) between them.

In softening results, TEGDMA/UDMA/0.1%CHX (G5) showed the higher value and it was not statistically different than the commercial infiltrant ($p>0.05$). Only the blends TEGDMA/0.1%CHX (G2), TEGDMA/0.2%CHX (G3) and TEGDMA/BisEMA/0.2%CHX (G9) were statistically different to the commercial infiltrant ($p<0.05$) and showed the lowest values of softening.

Concerning EM, all experimental resins had higher values and they were statistically different than the commercial infiltrant ($p<0.01$). TEGDMA/UDMA blends (G4, G5 and G6) and TEGDMA/BisEMA blend (G7) showed the highest EM values, followed by TEGDMA/BisEMA blends (G7, G8 and G9) and TEGDMA/UDMA blend (G4). TEGDMA blends (G1, G2 and G3) showed the lowest EM means. For FS, only TEGDMA blends (G1, G2 and G3) and the commercial infiltrant showed the lowest values and were not statistically different among each other ($p>0.05$). The other groups had higher FS and they did not show statistically significant differences among each other ($p>0.05$).

Table 3. Mean and standard deviation of sorption/solubility (µg/mm^3), softening (%), elastic modulus (GPa) and flexural strength (MPa) of experimental resin blends groups and commercial control group data.

Resin blends	**Sorption**	**Solubility**	**Softening**	**Elastic Modulus**	**Flexural Strength**
G1 (TEGDMA)	6.18 ± 0.51 **ab**	1.48 ± 0.31 **bc**	42.75 ± 4.75 **bc**	0.63 ± 0.04 **c**	48.00 ± 10.32 **b**
G2 (TEGDMA/0.1% CHX)	6.13 ± 0.37 **ab**	1.40 ± 0.42 **bc**	37.95 ± 12.35 **c**	0.66 ± 0.05 **c**	48.42 ± 4.64 **b**
G3 (TEGDMA/0.2% CHX)	6.34 ± 0.19 **a**	1.54 ± 0.09 **bc**	35.23 ± 8.12 **c**	0.63 ± 0.06 **c**	44.63 ± 8.97 **b**
G4 (TEGDMA/UDMA)	5.62 ± 0.55 **abc**	0.40 ± 0.24 **d**	43.67 ± 11.99 **bc**	0.83 ± 0.10 **ab**	77.65 ± 13.98 **a**
G5 (TEGDMA/UDMA/0.1% CHX)	5.54 ± 0.20 **bc**	0.53 ± 0.25 **cd**	57.92 ± 9.38 **a**	0.84 ± 0.06 **a**	75.73 ± 8.97 **a**
G6 (TEGDMA/UDMA/0.2% CHX)	5.33 ± 0.18 **c**	0.34 ± 0.17 **d**	44.52 ± 8.97 **bc**	0.85 ± 0.04 **a**	70.31 ± 9.31 **a**
G7 (TEGDMA/BisEMA)	3.73 ± 0.32 **d**	0.49 ± 0.25 **d**	42.96 ± 7.23 **bc**	0.80 ± 0.08 **ab**	74.16 ± 6.22 **a**
G8 (TEGDMA/BisEMA/0.1% CHX)	4.09 ± 0.38 **d**	0.61 ± 0.10 **cd**	43.41 ± 8.85 **bc**	0.73 ± 0.07 **bc**	72.90 ± 9.30 **a**
G9 (TEGDMA/BisEMA/0.2% CHX)	3.93 ± 0.10 **d**	0.67 ± 0.18 **bcd**	38.67 ± 6.01 **c**	0.73 ± 0.11 **bc**	68.59 ± 0.27 **a**
Icon®	3.54 ± 0.43 **d**	5.76 ± 1.15 **a**	51.70 ± 8.21 **ab**	0.43 ± 0.11 **d**	50.42 ± 12.78 **b**

Similar lowercase letters in column indicate no statistically significant difference between groups ($p>0.05$).

4. DISCUSSION

According to the results obtained in the present study, the characteristics of monomers caused different behavior of the experimental materials for all tests performed in this study; therefore the first hypothesis was accepted. The CHX addition was not the main responsible factor for the results, since monomers peculiarities were more relevant; the second hypothesis was reject, because 0.1 % of CHX influenced the TEGDMA/UDMA blends softening.

Water sorption is a condition that dental restorative materials are exposed to in the oral environment. The hydrolysis caused by water is an important factor to promote composite materials swelling and resin matrix degradation.[24] When a polymer is placed in water, hydrogen bonds are formed between water and polymer polar groups, as hydroxyl and carbonyl.[22] This condition can disrupt the polymer interchain interaction, such as entanglements and secondary bonding.[22] Water could then be considered as a solvent; the solvent effect on the polymer network is described as plasticization, situation that could alter the molecular structure and could increase the mobility of polymer chain segments.[22,23,26]

The changes caused by water can also occur because it has a small molecule size with high molar concentration.[29] Due to these characteristics, it can penetrate into nanometer size free volume spaces between polymer chains, or also it could keep itself linked with functional groups around by hydrogen bonding described above.[29] This behavior of water can cause modifications of the plasticization of polymer matrices, which compromise mechanical properties as yield deformation and, consequently, changes are reflected in reduction of resin materials mechanical properties.[22,29] The hydrophilic nature of polymers, associated with monomers chemistry characteristics and its polymerization linkages, could contribute for more or less plasticization during the water sorption.[30] On the other hand, it is known that conversion of double bonds in dental restorative materials is never totally completed because of reactions such as immobilization, gelatinization, vitrification or steric isolation that leaves pendant side chains, which act as permanent plasticizers.[31]

In this study, water sorption was not influenced by CHX presence or concentration, and could follow this order TEGDMA > UDMA > BisEMA and Icon®.

Similar results were shown by others that described this ranking of monomer water sorption TEGDMA > BisGMA > UDMA > BisEMA.[26,32] Although the resin blends of this study have as main constituent the dimethacrylate TEGDMA and is a high hydrophilic monomer, the addition of low percentage of different chemical chains as UDMA and BisEMA into TEGDMA resin blends interfered at the structure of polymer network.[26,33] TEGDMA blends tend to present denser network, but with non-homogeneous characteristics, that is associated with polymer bulk containing densely and loosely cross-linked.[26,33] This situation means microgels domain dispersed in a pool of unreacted monomers.[26,33] Therefore, this polymer chain interactions collaborates to high water sorption because the space created between the polymer clusters is larger and can accommodate more water.[26,33] Regarding BisEMA and UDMA, the presence of ether and urethane groups, respectively, make these monomers less hydrophilic.[33] BisEMA, when compared with its analogue BisGMA, is less hydrophilic because of the absence of hydroxyl groups, a characteristic that establish weaker hydrogen bond interactions with water molecules and promote higher resistance to degradation in the long term.[34]

Concerning solubility property, it is known that when some polymers are immersed in water, some of the components, such as unreacted monomers, are dissolved and leached from the samples.[22,26] In this study, Icon® showed high solubility, and the presence of initiators and stabilizers agents into the commercial infiltrant formulation could influence its higher capacity to solubilize in water environment than the experimental blends. In this study, TEGDMA neat blends were more soluble than the other experimental blends. This could be explained by TEGDMA's higher hydrophilic capacity than BisGMA and UDMA, and its weak molecular interactions.[35] In addition, the release of an amount of organic particles could be associated with the molecules size, and when the molecule is small, it has a tendency to enhance mobility, causing a faster elution than larger molecules.[24] If we compare the size of the monomers used in this study, TEGDMA has a smaller size than BisEMA and UDMA; this could have contributed to the high solubility of TEGDMA-neat blends. Nevertheless, chemical links obtained for polymerization and cross-link density seems to have more close association with the resin materials water solubility capacity.[24] These different resin blends behavior and the monomers capacity to uptake solvent could also be associated to the percentage of cross-link network between the chemical chains into the resin matrix.[27] The water sorption decreases when the cross-link increases.[22] On the other hand, water sorption increases with an increase in the hydrophilicity of the polymer network.[22]

In the present study, softening was obtained by KHN measurements. It is a method to evaluate softening ratio of resin materials based on repeated hardness measurements before and after the immersion of materials into ethanol.[36-38] Ethanol storage can cause a swelling, softening of resin materials surface and, consequently, leakage of unreacted monomer.[39] Thus, materials that have high amount of remaining double bonds tend to have more leakage, reducing the ethanol softening effect.[39] Moreover, higher cross-linked polymers are more resistant to degradation into solvent uptake challenge.[23,36] So, the softening ratio could be considered an indirect estimation for cross-link density.[16] For our results, the experimental infiltrants showed lower softening ratio than Icon®, except for the blend TEGDMA/UDMA/0.1%CHX that was similar to Icon®.

Concerning CHX presence, a study reported that 1% and 5% of CHX into some experimental BisGMA-based blends tested increased the degree of conversion, but decreased the elastic modulus property.[18] The presence of CHX in our blends did not interfere negatively in the softening of the most blends, that were better than the commercial control. Assuming that properties as degree of conversion and hardness could be directly correlated[40] the higher softening of TEGDMA/UDMA/0.1%CHX could be related to high KHN reduction for indirect calculation of softening. So, the presence of CHX in these blend specimens could be dissolved with the unreacted monomers, contributing for free spaces between the polymer bulk and consequently, decreasing the hardness.

The lower softening for the most blends could be happened because TEGDMA is the main constituent of all experimental blends. TEGDMA is a smaller molecule than UDMA and BisEMA, being a very flexible molecule with tendency to cyclization and to form cross-links.[27] Nevertheless, the pendant double bonds react to form primary cycles, which increase conversion, but do not contribute to network formation, leading to inhomogeneous polymer formation that could result in high polymerization shrinkage with potential gap formation within the cavity.[27] On other hand, it is known that the monomer UDMA contains an aliphatic spacer group between the methacrylates, and the presence of the urethane groups contribute to reactions at chemical chain, as soon as, the – NH – group contribute for chain transfer reactions, forming – N – radical.[25] This radical can cause the initiation of polymerization or cross-linking, decreasing the percentage of unreactive double bonds and optimizing the interchain interactions.[25] For this, TEGDMA-neat blends maybe are not favorable for the durability of an infiltrant material into fragile enamel porous because the higher hydrophilicity of TEGDMA could contribute for its susceptibility for hydrolyze.

In relation elastic properties as EM and FS, the viscosity of monomers could have influenced the resin materials tested; once the molecular interactions is related with strength that could influence to turn small molecules in larger structure.[30] Thus, the higher the viscosity, the higher is the resistance of molecules to flow and the higher is the molecular interactions.[25] Higher viscosity decreases the mobility of monomer during polymerization, decreasing flexibility of the corresponding polymeric network.[25] Short chains as TEGDMA represent molecular structure with low viscosity (0.05 Pa.s) and its molecular interactions are weaker than BisEMA (3 Pa.s) and than UDMA (8-10 Pa.s),[30,41] this difference of viscosities could influence the quality of elastic properties of our resin blends.

Regarding EM, Icon® showed the lowest values, followed by TEGDMA-neat blends. For FS, Icon® and TEGDMA-neat blends also showed the lowest means. The blends containing UDMA showed the highest values for EM and these blends were similar to BisEMA blends for FS. These results were in accordance to study[26] that showed EM of homopolymers following this order: TEGDMA < BisEMA < UDMA < BisGMA. Similar results to our study was described previously,[32] indicating that there is some relationship between FS and dymethacrylates homopolymers following this order: TEGDMA < BisGMA < UDMA. Concerning our results, the viscosity of each monomer could have influenced the elastic properties because the higher resistance strength was associated to the amount of double bonds converted and cross-link density, issues that have been already discussed above. In humidity conditions, FS and EM of polymeric matrix could be influenced by water uptake; water molecules have the potential to accommodate in micro-voids and keep inside the bulk by hydrogen bonds, forming clusters and do not cause swelling of polymer, acting as filler particles,[26] which could contribute to greater rigidity of the polymer.

Based on the these results and according to above discussions, characteristics as viscosity, chain size and reaction capacity of the monomers caused different behavior of experimental resin blends for the physical and mechanical properties evaluated. CHX did not influence most of the tests (sorption/solubility, elastic modulus and flexural strength) and it did not interfere negatively the softening of experimental materials (G1-G4; G6-G9), except for the blend G5 (TEGDMA/UDMA/0.1%CHX). Thus, CHX could be considered an option to add for an antimicrobial effect in infiltrant materials. These are the first results for experimental infiltrants with CHX-added and they could be considerable optimists concerning the tests evaluated. About the blends tested, the addition of UDMA in TEGDMA neat blend seems to be a promise, as soon as these blend showed less water sorption/solubility, which could influenced positively in a mouth environment.

5. CONCLUSIONS

The results of this study showed that the monomers chemical characteristics have more influence in the behavior of experimental blends than the antimicrobial additive. CHX in concentrations of 0.1% and 0.2% did not interfere with the physical-chemical properties evaluated, excepted for the softening test that TEGDMA/UDMA/0.1%CHX had the highest mean value. The experimental blends could be considered promising, as they performed better or equal to the commercial infiltrant, mainly UDMA containing blends, which showed the best results in the present study.

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2.2 ARTIGO 2

Three-body surface wear of experimental infiltrants chlorhexidine-added

ABSTRACT

The objectives of this study were to evaluate the effect of monomers in experimental resin blends CHX-added on three-body wear, and to verify the association between roughness measurements in 2D (Ra) and 3D (Sa) using a non-contact profilometer. Three experimental resin blends (TEGDMA/0.2%CHX; TEGDMA/UDMA/0.2%CHX and TEGDMA/BisEMA/0.2%CHX) and a commercial control infiltrant (Icon®) were evaluated (n=8). Blocks (5 mm x 3 mm x 2 mm) were prepared using a polyvinyl siloxane mold and the materials were light cured for 60 s. The resin blocks were clustered into acrylic resin and, after this, the clustered blocks were polished with sequence of grits. The parallelism of clustered blocks surfaces were checked with caliper. Then, the measurements of blocks surface and initial roughness in 2D (Ra) and 3D (Sa) were measured by non-contact profilometer. Three-body wear was accomplished by V8 toothbrush prototype device (10.000 cycles with load of 350 gF) with soft toothbrushes and medium abrasive dentifrice (slurry 1:1 with deionized water). After toothbrushing assay, the final roughness (Ra and Sa) and material loss were evaluated. Roughness data were analyzed using ANOVA for repeated measures and Tukey test (α=5%). For material loss one-way ANOVA and Tukey test (α=5%) were applied. The association of Ra and Sa roughness was verified using simple linear regression. After toothbrushing, TEGDMA/UDMA/0.2%CHX blend showed the lowest values for roughness (Ra and Sa) and material loss, and it was different than control ($p<0.01$). The blend TEGDMA/BisEMA/0.2%CHX had the worst results with the highest values of roughness and material loss. There was statistically significant association ($p<0.01$) between Ra and Sa roughness (R^2=74.56%). The materials wear was influenced by resin blends monomeric composition and they were similar or better than Icon®. The two methods of roughness evaluation (2D and 3D tests) showed satisfactory association between them.

KEY WORDS: Chlorhexidine, infiltrants, three-body wear, toothbrushing, superficial roughness.

1. INTRODUCTION

Minimal intervention dentistry is an approach that aims to keep teeth functional tissues through earliest dental caries detection, prevention the disease progression and by ultraconservative restorative treatments (Kielbassa et al., 2009; Leal, 2014; Jingarwar et al., 2014). This concept of maximum preservation of remaining dental structures can be applicable in every dental specialty, particularly in pediatric dentistry (Leal, 2014). Between the less invasive techniques for dental caries intervention, the infiltration of early caries lesions is a promising method to control and even to arrest the lesion progression by penetration of demineralized porous enamel with low viscosity light curing resin (Garcia-Godoy et al., 1997; Paris et al., 2007; Meyer-Lueckel and Paris, 2008). The infiltrant material and resin blends with high amount of triethylene glycol dimethacrylate (TEGDMA) are capable to this (Paris et al., 2007; Meyer-Lueckel and Paris, 2008).

Currently, a TEGDMA based-material is commercially available (Icon®, DMG, Hamburg, Germany). Studies *in vitro* (Paris et al., 2012; Torres et al., 2011; Oliveira et al. 2015), *in situ* (Paris and Meyer-Lueckel, 2010) and clinical trials (Paris et al., 2010; Meyer-Lueckel et al., 2012) have been accomplished to evaluate the performance of this material and all of them have been showed favorable results concerning penetrability capacity to arrest initial caries lesions, as soon as satisfactory performance masking white spots and controlling enamel erosion. Nevertheless, some studies have shown results that make questionable the real efficiency of commercial infiltrant. A randomized clinical trial of three years showed that Icon® and a nanoparticulate adhesive system fluoride-release had not showed statistically significant difference arresting early caries lesions progression in proximal surfaces of adult patients (Martignon et al., 2012). Fact that can favorable others techniques such as the use of fluoride, application of silver diamine fluoride and sealants, which could spend less time for application than the infiltrant technique, principally in a pediatric service, which clinical proceeding need to be fast (Leal, 2014; Mattos-Silveira et al., 2014). The capacity to decrease bacterial growth of infiltrated enamel surface with Icon® was also evaluated and the infiltrated surface showed more *Streptococcus mutans* adhesion than an enamel surface treated with varnish (Arslan et al., 2015). No relate was observed about an antimicrobial component in Icon® formulation. Therefore, the addition of an antimicrobial agent could be a promise propose to improve infiltrant benefits, avoiding biofilm adhesion over treated areas.

In front of some questionable points about Icon®, studies have been tested experimental resin blends with infiltrant characteristics with intention to find the best resin formulation capable to fill the gaps founded in commercial infiltrant (Araújo et al*.,* 2013; Araújo et al., 2015). Then, experimental infiltrants with TEGDMA, UDMA (urethane dimethacrylate) and BisEMA (bisphenol A ethoxylated dimethacrylate) based blends, without diluent as ethanol or HEMA (2-hydroxyethyl metacrylate), seemed to be promising blends for infiltrant functions (Araújo et al., 2013; Araújo et al., 2015).

Concerning the bacterial growth over the infiltrated surface, the addition of an antimicrobial in these experimental infiltrants could inhibit biofilm growth over and around the infiltrated area, and the chlorhexidine diacetate (CHX) could be an alternative for this. The addition of low concentration of CHX in dental materials as resin blends and glass ionomer cements was related without considerable changes in material properties, and could be an alternative to inhibit biofilm aggregation (Takahashi et al., 2006; Cadenaro et al., 2009; Hiraishi et al., 2010; Stanislawczuk et al., 2014). However, it is known that at oral cavity, the resin dental materials are susceptible to various chemical and mechanical challenges and CHX do not present chemical links with polymer chains, staying entrapped into spaces of polymer bulk (Riggs et al., 2000; Anusavise et al., 2006). In humid conditions, the CHX and unreacted monomers could be susceptible to be leach from the resin to the mouth environment, decreasing materials properties (Leung et al., 2005; Hiraishi et al., 2008).

Thereby, the resistance against adhesive, abrasion, fatigue and corrosion wear represents an important factor to predict the clinical success of restorations (Turssi et al., 2003). It is certain that oral challenges promoted by mastication of different foods, drinks and the contact with saliva and microorganisms can decrease the efficiency of restorative materials (Turssi et al., 2003). Simulation of toothbrush with dentifrice could also be used to evaluate the surface texture and wear abrasion of dental resin materials, as soon as, the corrosive wear is a continuous process occurring during the lifetime of restorations (De Paula et al., 2011). For the better or the worst wear resistance, the composition of resin materials can be influenced by monomers and diluents with their different molecular structures and chemical characteristics, factors like these are directly associated with important properties as hardness, cross-link density, higher conversion and hydrolytic capacity (De Paula et al., 2011; Carvalho et al., 2012).

For surface quality evaluation, the use of profilometer has been the most common method concerning roughness measurements in Ra (Takahashi et al., 2013). The non-contact profilometer method could be an alternative to measure superficial rough. The high sensitivity of 3D images method, that show rough measurements in Sa, were more precise than conventional profilometer for superficial roughness measurements over dental materials after toothbrushing because its more surface details (Garcia-Godoy et al., 2009). Nevertheless, it could be interesting to verify if conventional rough measurements in Ra could be associated with roughness measurements in Sa, predicting the values each other. The existence of some association could corroborate with rough evaluation in Ra, as soon as 3D images measurements depend of very specific devices.

Thus, the objectives of this study were to evaluate the effect of monomers in experimental resin blends CHX-added on three-body wear, and to verify the association between roughness measurements in 2D (Ra) and 3D (Sa) using a non-contact profilometer. For this study, the first tested hypothesis was that mechanical degradation could affect the roughness and material loss of experimental infiltrants and the second hypothesis was that roughness measured in 2D and 3D could be associated.

2. MATERIAL AND METHODS

2.1. Experimental resin blends preparation

For this study, three experimental resin blends with infiltrant characteristics were prepared (Table 1). These experimental materials consisted as their mainly constituent the high fluid monomer TEGDMA (Sigma-Aldrich, St. Louis, MO, USA, Batch STBD4587V). Dimethacrylates as UDMA (Sigma-Aldrich, St. Louis, MO, USA, Batch MKBK4814V) and BisEMA (Sigma-Aldrich, St. Louis, MO, USA, Batch MKBR5994V) were added in other two resin blends, but in less percentage (wt/wt).

The photoinitiator system used in these experimental resin blends were 1.0 wt% of DMAEMA (2-dimethylaminoethyl methacrylate, Alfa Aesar, Lancashier, United Kingdom, Batch 10155620), 0.5 wt% of camforquinone (Sigma-Aldrich, St. Louis, MO, USA, Batch 09003AQV) and 0.1 wt% of BHT (butylated hydroxytoluene, Sigma-Aldrich, St. Louis, MO, USA, Batch MKBQ2106V) was used as inhibitor agent. A concentration of 0.2% of CHX (chlorhexidine diacetate salt hydrate, Sigma-Aldrich, St. Louis, MO, USA, Batch SLBF1787V) was added in the three resin blends. All chemical components were weighed, separately, using an analytical balance (AL204, Mettler Toledo, Columbus, OH, USA). The blends were prepared at room temperature in a dark environment, mixing all the components of each blend using a spatula. After this, each final blend was stored in clean and dry amber glass bottles, in a dark environment at 4°C to avoid the polymerization process with the intention to keep all the blends in good conditions to use. As commercial control group the infiltrant Icon® (DMG – Hamburg, Germany, Batch 707408) was used.

Table 1. Composition of low viscosity monomer blends with infiltrant characteristics used in this study and commercial control group.

Groups	Composition
G1	TEGDMA (99.8 wt.%), CHX 0.2 wt.%
G2	TEGDMA (74.8 wt.%), UDMA (25 wt.%), CHX 0.2 wt.%
G3	TEGDMA (74.8 wt.%), BisEMA (25 wt.%), CHX 0.2 wt.%
Icon®	TEGDMA-based resin, initiators and stabilizers

2.2. Preparation of resin blends samples

For this step, rectangular samples (n=8) with dimensions of 5 mm x 3 mm x 2 mm (length x width x high) were prepared for experimental infiltrants and Icon®, using a polyvinyl siloxane mold (Aquasil Ultra Rigid, Dentsply, York, PA, USA). After filling the mold rectangular cavity with resin blend, a glass microscope slide (Fisher Scientific, Pittsburgh, PA, USA) was put covering the set before the light curing process to obtain a flat and smooth surface. The absence of bubbles was verified inside the resin blend in the mold. Thereafter, the sample were light cured for 60 s with LED Elipar S10 (3M ESPE, St. Paul, MN, USA) with power density of approximately 1000 mW/cm^2. The resin blends samples were kept in 100% relative humidity environment for 24 h at 37°C.

For resin blends samples engagement in the brushing machine, the samples needed to be clustered into acrylic resin denture base (Repair Material powder/liquid, Dentsply, York, PA, USA). A rectangular teflon mold with dimensions of 19 mm x 12.7 mm x 8.26 mm (length x width x high) was used to cluster the resin samples. Therefore, the resin samples were clustered in a central region of teflon mold to place them in contact with the central portion of toothbrush bristles during the brushing assay. After this, the clustered samples, named resin blocks after now, were submitted to polish process to standard the surface before brushing assay.

2.3. Standardization and polishing method of resin blends samples

The resin blocks surface was polished following the ISO 11609 (2010) recommendations. The polish machine AutoMet 250 (Buehler, Lake Bluf, IL, USA) was used to polish all resin blocks with rotation speed of 50 rpm and vertical load of 6 lbs (26.69 N). The sequence of grits (Buehler, Lake Bluf, IL, USA) used to remove resin beads and to leave smooth the blocks superficial surface was 240, 400, 600, 800 and 1200, one minute each. After this, a diamond paste system (disk + paste) 3MIC and MetaDi 1.0 µm (Buehler, Lake Bluf, IL, USA) was used following by the Trident fiber pad (Buehler, Lake Bluf, IL, USA), also one minute each disk. Running tap water was used to rinse the blocks after polishment.

About the blocks shape evaluation, it was checked measuring the parallelism of superior and inferior surfaces in four different directions: longitudinal, transversal and the two diagonals. A caliper (Brown & Sharpe, Stockholm, Sweden) was used to measure the heights between the two surfaces, and a delta height value obtained (greater height value - smaller height value). The resin blocks that showed delta values greater than 0.1 mm had to be repolished and the height delta value measured again.

After all the resin blocks passed in parallelism surfaces requirements, the scanning using the profilometer Taylor Robson CCI (Ametek, Chigago, IL, USA) was processed. The preparation of blocks surfaces until this step had to be accomplished because the profilometer realize a non-contact surface reading with emissions of wavelengths, and any level gap on surface can cause error in the final reading.

In this study, a magnification lens of 5 X, with numerical aperture of 0.13 mm were used for all surfaces scanning by profilometer. For superficial roughness, the profilometer software was set up to take 2D and 3D measurements, which means values of roughness were expressed in Ra for 2D images by three equidistant lines measurements over the resin specimens using Gaussian filter (cut off) of 0.8 mm and in Sa for 3D images by full specimen surface scanning. About the requirements to measure the rough, the profilometer had received manufacture calibration to offer precise measurements according to ISO for geometrical product specifications (ISO 4287 for Ra and ISO 4287 for Sa).

2.4. Sample selected by initial Ra and Sa roughness determination

The profilometer was used to evaluate more two requirements before start the toothbrushing assay: the level between acrylic resin and resin blend specimen, and the initial roughness of resin blend specimen. For this, the profilometer software was set up according to resin blend dimensions and limit measurements were defined to consider the final specimen as “passed” and “failed” for toothbrush assay. The relevant values of Ra for specimens requirements were used as initial roughness for Ra and the same scanned image was used to obtain Sa initial roughness.

2.5. Three body wear

After specimen’s standardization they were submitted to mechanical degradation. For this, an adhesive tape (Argon Inc., Monrovia, CA, USA) with a pre-fabricated rectangular window (3 mm x 6 mm) was positioned over the central region of specimen and perpendicular of the greater size of specimen. The toothbrush was conducted by toothbrush device V8 model (prototype device of Bioscience Laboratory of University of Tennessee Health Science Center) realizing 2.78 strokes/second, with a load of 350gF at each toothbrush, for 10.000 cycles of brushing, approximately 1 year of degradation (Garcia-Godoy et al., 2009). The toothbrushes with soft bristle (ADA, Chicago, IL, USA) were kept at 100% of absolute humidity (deionized water) for 24h at room temperature before start the toothbrushing. The slurry was prepared using a popular dentifrice of medium abrasion (Colgate Cavity Protection[3], Colgate-Palmolive Company, New York, NY, USA), in a proportion of 1:1 for deionized water. Thus, 37.5 g of water and 37.5 g of dentifrice were weighed using a precision balance (ML, Mettler Toledo, Columbus, OH, USA) (Garcia-Godoy et al., 2009). After the toothbrushing cycles, the final roughness in Ra and Sa were measured for all specimens using the same process used for initial roughness at profilometer. Nevertheless, it is important to attempt that the final Ra measurements was did in opposite direction of brushing wear.

2.6. Material loss measurement

The material loss was calculated by the difference (in µm) between the mean heights (Z) of brushed region and non-brushed region. The scanning of specimen area was taken by profilometer and this area was divided in three parts: brushed area (region 1) and two no-brushed areas (region 2). The profilometer software processed the values using the following equation:

$$\textit{Height difference} = Z \textit{ mean } (2) - Z \textit{ mean } (1)$$

2.7. Statistical analysis

The statistical analysis for Ra and Sa roughness values were obtained by ANOVA with repeated measures and Tukey test (α=5%). In case of material loss after toothbrushing assay, one-way ANOVA with Tukey test (α=5%) was used to evaluate the data. Then, to compare possible association between Ra and Sa roughness measurements, a simple linear regression was accomplished. The SAS System release 9.3 (SAS Institute Inc., Cary: NC, 2012) was used in all statistical analysis.

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[3] **Colgate Cavity Protection composition**: **a) Active ingredient:** Sodium monofluorophosphate 0.76% (0.15%w/v fluoride ion); **b) Inactive ingredients:** dicalcium phosphate dihydrate, water, glycerin, sodium lauryl sulfate, celulose gum, flavor, tetrasodium pyrophosphate, sodium saccharin.

3. RESULTS

3.1. Ra roughness

For Ra roughness, the interaction of treatment (groups) and phase (before and after mechanical degradation) was statistical significant ($p<0.0001$). According to table 2, all experimental blends and Icon® had difference of roughness before and after mechanical degradation. In initial roughness value, the medium values did not show significant statistical difference for all groups, and their surfaces were smoother than after degradation. Concerning the values of roughness after degradation, the group TEGDMA/BISEMA/0.2%CHX showed the higher surface roughness when compared with the other groups. The group TEGDMA/UDMA/0.2%CHX was the lowest roughness after degradation and it was not different than Icon® roughness before toothbrushing.

Table 2. Mean ± standard deviation of roughness (Ra) of experimental resin blends and commercial control group.

Groups	Phase			
	Initial roughness (Ra)		**Final roughness (Ra)**	
TEGDMA/0.2%CHX	0.0027 ± 0.0011	aB	0.0107 ± 0.0046	bA
TEGDMA/UDMA/0.2%CHX	0.0023 ± 0.0007	aB	0.0062 ± 0.0011	bA*
TEGDMA/BisEMA/0.2%CHX	0.0033 ± 0.0003	aB	0.0241 ± 0.0039	aA
Icon®	0.0043 ± 0.0021	aB*	0.0095 ± 0.0054	bA

Capital letters (line): same letters are not statistically significant difference between each other;
Small letters (column): same letters are not statistically significant difference between each other;
Symbol (*): no statistically significant difference between each other;

3.2. Sa roughness

About Sa roughness (Table 3), the statistical analysis showed differences ($p<0.0001$) between the treatments (infiltrant groups) and between phase (before and after mechanical degradation). As soon as the interaction between treatment and phase was also statistical significant ($p<0.0008$). The roughness values were higher after degradation than before degradation for all groups. Concerning initial roughness, the groups showed significant statistical difference when they were compared each other and TEGDMA/BisEMA/0.2%CHX was the most rough, following by TEGDMA/0.2%CHX and Icon®. TEGDMA/UDMA/0.2%CHX was the least rough before degradation. After toothbrushing, the roughness values increased expressively and TEGDMA/BisEMA/0.2% and TEGDMA/0.2%CHX showed the higher roughness values, following by Icon®. The group TEGDMA+UDMA+0.2%CHX was the less rough after degradation and it was not different than Icon® roughness before toothbrushing. Icon® roughness after degradation was similar to TEGDMA/0.2%CHX before toothbrushing.

Table 3. Mean ± standard deviation of roughness (Sa) of experimental resin blends and commercial control group.

Groups	Phase			
	Initial roughness (Sa)		Final roughness (Sa)	
TEGDMA/0.2%CHX	0.5486 ± 0.1385	bB$^{\alpha}$	2.0676 ± 0.8421	bA
TEGDMA/UDMA/0.2%CHX	0.0967 ± 0.0200	dB	0.1934 ± 0.0204	dA*
TEGDMA/BisEMA/0.2%CHX	0.8641 ± 0.1125	aB	3.4700 ± 0.7285	aA
Icon®	0.2575 ± 0.0323	cB*	0.5790 ± 0.1047	cA$^{\alpha}$

Capital letters (line): same letters are not statistically significant difference between each other;

Small letters (column): same letters are not statistically significant difference between each other;

Symbols (α, *): same symbols are not statistically significant difference between each other.

3.3. Material loss after toothbrushing

According to Table 4, it could be observed significant statistical difference between the material loss for all groups (p<0.0001). Therefore, TEGDMA/BisEMA/0.2%CHX group showed the highest material loss. TEGDMA/0.2%CHX and Icon® presented intermediary values and TEGDMA/UDMA/0.2%CHX was the resin blend that showed the most resistance after degradation process.

Table 4. Mean ± standard deviation of material loss (µm) of experimental resin blends and commercial control group.

Groups	Material loss (µm)	
TEGDMA/0.2%CHX	4.98 ± 0.98	b
TEGDMA/UDMA/0.2%CHX	0.45 ± 0.16	d
TEGDMA/BisEMA/0.2%CHX	7.89 ± 0.98	a
Icon®	0.89 ± 0.36	c

Different lowercase letters indicates statistically significant difference between groups.

3.4. Association between Ra and Sa roughness measurements

The linear regression was applied using a conceptual model (y = a+b.x), where Ra was defined as predict variable and Sa as answer variable. So, according to Figure 1 , from roughness data, the results of regression study analyzed by statistical program created the following equation, that it represents a **R^2 value of 74.56% with statically significant true association ($p<0.01$):

$$\boldsymbol{Sa\,roughness = -0.02491 + 131.21747 * Ra\,roughness}$$

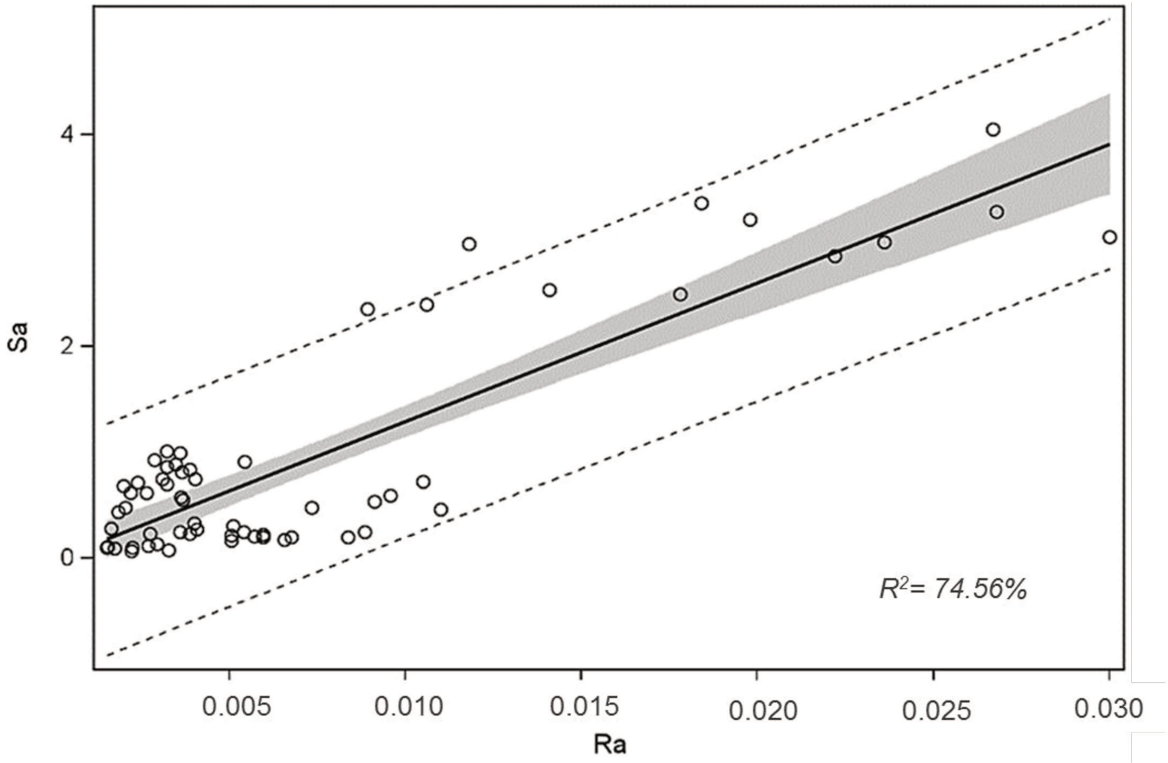

Figure 1. Linear regression of roughness in Sa in function of roughness in Ra.

4. DISCUSSION

The present study was conducted to evaluate the effects of different monomers into experimental resin blends and a commercial control after toothbrushing challenge. It was observed that superficial roughness, both for 2D and 3D evaluations, increase according to formulation of resin materials. The same occurred for material loss, where BisEMA was less resistant to toothbrushing challenge. Thus, the first tested hypothesis could be considered accepted. BisEMA based-infiltrants presents the highest roughness, followed by TEGDMA resin blend, commercial control and the UDMA was the lowest rough. These results could be associated to polymerization degree of monomers at polymer bulk. As soon as the lower degree of polymerization could cause less wear resistance due to poorer physical properties (Kawai et al., 1998). In a study that tested experimental resins, the wear resistance of UDMA/TEGDMA resins increased by increased the TEGDMA and UDMA contents and these blends had higher resistance than BisGMA-based composites. In this same study, the authors found an inversely proportional relation between surface hardness and amount of wear for experimental resins tested (Kawai et al., 1998).

Considering that higher values of hardness could influence the wear resistance, higher Knoop hardness was showed for experimental infiltrants based on TEGDMA, UDMA and BisEMA methacrylates when they were compared to Icon® (Araújo et al., 2013). The blends formulated with 100% of TEGDMA, 75% of TEGDMA and 25% of UDMA, 75% of TEGDMA and 25% of BisEMA were not statistical different to Icon® and showed Knoop hardness number higher than two times when compared with commercial infiltrant. Due to these previous results, the present study was designed and the results could collaborate to explain the bad performance data obtained concerning BisEMA resin blends. Thus, the addition of BisEMA in TEGDMA blends could collaborate to decrease conversion of double bonds because BisEMA has high molecular weight that due less flexible the monomer chemical chain during polymerization process (Ferracane, 2006; Van Landuyt et al., 2007). On the other hand, UDMA has a small size and low viscosity than BisEMA, which contribute for high amount of double bonds (Sideridou et al., 2003). Furthermore, the addition of UDMA into TEGDMA blend could corroborate to increase mechanical properties as degree of conversion, elastic modulus and Knoop hardness by great performance of polymerization (Araújo et al., 2013).

About the data of commercial control group, it was showed the second better wear resistance. It is known that composition of this material consist in aliphatic dimethacrylates, and it has TEGDMA as main constituent, according to manufacturer. The TEGDMA presents high capacity to convert double bonds because it is very fluid and has a small size of chemical chain, contributing for more flexibility to convert double bonds (Van Landuyt et al., 2007). The Icon® toothbrush wear resistance performance was compared with an adhesive system in infiltrated artificial white spot lesions using bovine teeth and a toothpaste with high abrasive potential (Belli et al., 2011). Although no statistical differences were found between both materials, the authors observed that infiltrated areas with the adhesive system were more heterogeneous and had less depth penetration with exposition of unfilled surface underneath, fact that could be associated with a tendency to have more surface degradation over time (Belli et al., 2011). However, Icon® showed more homogeneous depth of penetration with a tendency to improve the quality stability of material that could be associated to the ethanol drying protocol application and for the TEGDMA superior infiltration ability in water free surfaces (Belli et al., 2011).

Although the good wear resistance of Icon®, it not showed the better superficial rough even after systematic polishment. The surface rough of infiltrated enamel with Icon® has been reported for previous studies (Muller et al., 2011; Yang et al., 2012). The infiltrant layer on enamel surface is non-homogeneous after light curing, and irregularities and excesses of resin could be favorable to biofilm formation, especially in proximal surfaces (Muller et al., 2011; Yang et al., 2012). Even using abrasive strips after light curing, this procedure did not show improvement of smoothness of infiltrated enamel; and removal of excess material before light curing could be recommendable to avoid unexpected abrasion resulting from polish strips (Muller et al., 2011; Yang et al., 2012). This concern about rough surface of resin materials which is placed over the teeth is important, as soon as it is known that a rough greater than 0.2 µm is favorable for biofilm development (Bollen et al., 1997). So, it is prudent to observe, in the present study, that the values of Sa (µm) for resin materials tested, excepted to TEGDMA/UDMA/0.2%CHX, were considerable higher than 0.2 µm after toothbrush. The less rough of TEGDMA/UDMA/0.2%CHX even after wear abrasion could be a positive characteristic for this experimental infiltrant material because the constant biofilm oral challenges conditions.

Concerning the 2D and 3D measurements for superficial roughness, the non-contact profilometer was used. This methodology was more sensitive than contact roughness device (Garcia-Godoy et al., 2009). Furthermore, in the present study, it was found that the values in Ra and Sa showed satisfactory association (R^2=74.56%) between both measurements, and the second hypothesis was accepted. This fact represents that Ra and Sa are numerical quantities associated and in our results for roughness, Sa showed more differences between the groups tested, supposing that Sa could be more precise than Ra. This could be happened because 2D parameters does not contain any information about superficial textural characteristics, limiting roughness values evaluation and it being considerable a simplify method (Mueller et al., 2011). In contrast, Sa has a possibility to evaluate the superficial area by arithmetic average of the 3D roughness and it has being considerable a more representative way to measure surfaces textures (Mueller et al., 2011; Ulrich et al., 2014). Another advantage is that 3D non-contact profilometer makes possible to measure the wear depth and roughness of the worn surface consequently using the same instrument, avoiding possible measurements errors by scratches presence over the specimens when contact device is used (Cao et al., 2013).

Regarding the clinical relevance of this study, to know about wear resistance of infiltrant materials after mechanical degradation is important since in a clinical situation, these materials will be susceptible to challenges of mouth environment including the various periods of toothbrush with toothpaste. In front of the results obtained from this study, the experimental infiltrants with CHX added can be considered with high potential to be an infiltrant product because they are similar or better than a commercial control. Nevertheless, the presence of different methacrylates added in TEGDMA seems to interfere at wear resistance of experimental materials after toothbrush challenge. The blend containing the viscous methacrylate UDMA showed the high performance after toothbrush cycles, being more resistant for this *in vitro* simulation. However, more studies need to be accomplished for better characterized these experimental blends until introduce new proposals for the dentistry community.

5. CONCLUSION

Based on the conditions of the present study it was possible to conclude that the degradation of materials evaluated was influenced by monomer based composition and the experimental resin blends were similar or better than commercial infiltrant. The two methods of roughness evaluation (2D and 3D tests) showed satisfactory association between them.

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3. DISCUSSÃO

O uso de um material com capacidade infiltrante na paralisação de lesões iniciais de cárie tem sido considerado uma técnica eficiente desde que o infiltrante Icon® foi lançado comercialmente (Paris et al., 2007; Meyer-Lueckel et al., 2011). Devido à grande repercussão quanto aos conceitos atuais de mínima intervenção para os tratamentos restauradores, técnicas não-invasivas e micro-invasivas passaram a ser fortemente investigadas, melhoradas ou desenvolvidas (Leal, 2014; Jingarwar et al., 2014). Apesar do conceito de infiltrar superfícies porosas como o esmalte desmineralizado utilizando-se material resinoso ser bastante antiga (Dávila et al., 1975; Robinson et al., 1976), há estudos recentes que contribuíram consideravelmente para que o conceito de infiltração fosse estabelecido na comunidade científica Odontológica (Paris et al., 2007; Meyer-Lueckel et al., 2007; Meyer-Lueckel e Paris, 2008; Meyer-Lueckel e Paris, 2010; Paris et al., 2010). Atualmente, o Icon® é o único material infiltrante disponível comercialmente no mercado Odontológico. A capacidade de penetração deste material nas lesões naturais de cárie, preenchendo eficientemente o corpo da lesão de mancha branca foi mostrado por Meyer-Lueckel et al., (2011), comprovando que o dimetacrilato TEGDMA, principal constituinte do Icon®, seja uma escolha apropriada para a formulação de materiais com alto poder de penetração.

Esta tese, dividida em dois capítulos, objetivou avaliar as propriedades físico-químicas de nove misturas resinosas experimentais com características infiltrantes, sendo que duas concentrações de diacetato de clorexidina (0,1% e 0,2%) foram adicionadas em parte das misturas com o intuito de promover ação antimicrobiana aos materiais experimentais. No Capítulo 1, foram realizados os testes de sorção e solubilidade, taxa de redução de dureza, resistência à flexão e módulo de elasticidade. Essas propriedades podem estar relacionadas com a capacidade de absorver água e eliminar partículas por meio da solubilidade do polímero (Capítulo 1). Já no Capítulo 2, os materiais experimentais com adição de 0,2% de CHX foram submetidos ao desgaste mecânico por três corpos utilizando um dentifrício de média abrasão, simulando um período de escovação de aproximadamente 1 ano. A presença de diferentes monômeros como TEGDMA, UDMA e BisEMA influenciou a resistência à degradação (Capítulo 2).

Em relação à composição dos infiltrantes experimentais avaliados no presente estudo, a escolha pelos monômeros dimetacrilatos TEGDMA, UDMA e BisEMA foi baseada em estudos prévios (Araújo, 2011a; Araújo, 2011b; Sfalcin 2011). Esses pesquisadores observaram que essas misturas seriam promissoras para um material com características infiltrantes. Araújo (2011a) avaliou propriedades físico-mecânicas de infiltrantes experimentais comparados ao infiltrante comercial (Icon®) e observou que misturas à base TEGDMA e/ou UDMA, sem a adição de diluentes como o HEMA e etanol, foram semelhantes ou melhores que o controle comercial em relação ao grau de conversão, densidade de ligações cruzadas, módulo de elasticidade e dureza Knoop. O monômero BisEMA influenciou negativamente nas propriedades analisadas para todos os testes avaliados. Araújo (2011b) também mostrou que os infiltrantes experimentais sem a adição de diluentes apresentaram melhor resistência de união após a infiltração em esmalte cariado. O grau de penetração desses infiltrantes experimentais em lesões subsuperficiais em esmalte foi avaliado por Sfalcin (2011) e todas as misturas foram consideradas com boa capacidade de penetração dentro do corpo da lesão. No entanto, a mistura contendo 75% de TEGDMA e 25% de BisEMA, sem a adição de diluentes, apresentou maior grau de penetração.

Para promover capacidade antimicrobiana aos infiltrantes experimentais citados acima, adição das duas concentrações de CHX às misturas experimentais foi proposto em um estudo anterior (Inagaki, 2012). As misturas contendo CHX apresentaram atividade antibacteriana eficiente, uma vez que apresentaram Concentração Mínima Bactericida (CMB) de $9,7 \times 10^{-4}$ para *Streptococcus mutans* e *Lactobacillus acidophilus*. Após teste de difusão em ágar (Pour Plate), as misturas polimerizadas apresentaram ação antimicrobiana apenas para *S. mutans* e a mistura contendo 75% de TEGDMA, 25% de UDMA e 0,1% de CHX mostrou o maior halo de inibição. O infiltrante Icon® não apresentou atividade antibacteriana em nenhum dos testes conduzidos. Apesar de representarem resultados iniciais os resultados acima descritos podem ser considerados otimistas, pois mostraram a eficiência da CHX em proporções muito pequenas e principalmente, mostrou capacidade de atuação contra *S. mutans* após a polimerização.

Além do teste microbiológico, também foram avaliados, propriedades físico-mecânicas como grau de conversão e dureza Knoop superficial dos infiltrantes experimentais com adição de CHX e estes foram comparados ao Icon® (Inagaki, 2012). A concentração de CHX não afetou as duas propriedades avaliadas, entretanto, o tipo de base monomérica em cada formulação influenciou as propriedades avaliadas. As misturas à base de 75% de TEGDMA e 25% de UDMA, independente da concentração de CHX, mostraram os melhores resultados em relação aos testes executados.

A incorporação de CHX aos materiais restauradores não afetou propriedades como grau de conversão e módulo de elasticidade em adesivos experimentais à base de TEGDMA e BisGMA contendo CHX a 1%. A incorporação de CHX a 1% em cimentos de ionômero de vidro não afetaram propriedades como resistência à compressão e resistência de união (Takahashi et al., 2006), e em sistemas adesivos, não influenciou propriedades como a microtração (Hiraishi et al., 2010). Esse resultado corrobora com os observados no presente estudo. Além disso, é importante salientar que se utilizou concentração 5 vezes maior que a apresentada nesta tese (Cadenaro et al., 2009).

De acordo com os resultados obtidos no Capítulo 1, as diferentes concentrações de CHX (0,1% e 0,2%) não influenciaram as propriedades de sorção e solubilidade dos materiais testados, sendo as características monoméricas das misturas responsáveis pela maior ou menor capacidade de captação de água do polímero. Quando se trata de compósitos, a matriz orgânica é dependente da estrutura química, densidade de ligações cruzadas e grau de heterogeneidade molecular do polímero final (Pfeifer et al., 2011). Além disso, fatores como o grau de conversão e resistência à degradação em solventes orgânicos estão diretamente relacionados com a durabilidade do material, uma vez que os desgastes químicos e mecânicos são inevitáveis na cavidade bucal (Heintze et al., 2007; Pfeifer et al., 2011).

Os infiltrantes experimentais avaliados neste presente estudo possuíam o TEGDMA como monômero-base. O TEGDMA é um dimetacrilato de baixo peso molecular e bastante fluido, com alto poder plasticizante e por isso, muito utilizado como diluente na composição de resinas compostas e sistemas adesivos (Van Landuyt et al., 2007; Pfeifer et al., 2011). O TEGDMA possui uma cadeia alifática bastante flexível, em que os grupos éteres atuam como receptores para ligações de hidrogênio (Sideridou et al., 2003; Pfeifer et al., 2011). Entretanto, a presença dessas ligações éteres contribui para que o TEGDMA apresente características hidrofílicas maiores que o UDMA e o BisEMA (Ferracane et al., 2006). Fato que pode explicar os maiores valores de sorção e solubilidade encontrados para as misturas contendo apenas TEGDMA na composição.

No entanto a adição de UDMA e BisEMA às misturas à base de TEGDMA proporcionou mudanças no desempenho dos materiais. Essas misturas apresentaram capacidade reduzida de sorção de água e solubilidade dos materiais e aumentaram a resistência à flexão e o módulo de elasticidade (Capítulo 1). A razão módulo de elasticidade/dureza de superfície é proporcional ao índice de plasticidade do material, ou seja, quanto maior a razão, melhor será a capacidade do material suportar microtrincas causados por agentes plastificantes (Heintze et al., 2007). Este princípio poderia estar relacionado com o desempenho das misturas experimentais em relação às propriedades avaliadas como o módulo de elasticidade e resistência ao desgaste mecânico. Misturas contendo apenas TEGDMA apresentaram os menores valores de módulo de elasticidade, sendo a mistura que apresentou considerável perda de estrutura após teste de escovação. Já no Capítulo 2, a mistura que continha UDMA foi a mais resistente à degradação mecânica, mostrando menor rugosidade e menor perda de material. No entanto, a mistura que continha BisEMA mostrou ter a menor resistência ao desgaste mecânico.

O monômero BisEMA é um análogo do BisGMA, possuindo dois anéis aromáticos que contribuem para sua menor flexibilidade durante o processo de polimerização, convertendo menos ligações duplas (Van Landuyt et al., 2007; Sideridou et al., 2003), fato este que pode ter interferido na menor resistência ao desgaste mecânico. Em relação ao BisGMA, o BisEMA é menos viscoso e possui menor capacidade de sorver água pela ausência dos radicais hidroxilas (Ferracane, 2006; Sideridou et al., 2002), justificando a menor sorção observada no Capítulo 1. Já o UDMA possui viscosidade maior que o TEGDMA e o BisEMA o que indica presença de ligações intermoleculares mais intensas, fazendo que o monômero seja menos flexível durante a polimerização quando comparado ao TEGDMA (Sideridou et al., 2002; Sideridou et al., 2003). A presença dos radicais uretanos faz com que ele tenha capacidade de formar ligações cruzadas mais fortes, proporcionando matrizes poliméricas mais densas (Sideridou et al., 2003). No entanto, as misturas contendo UDMA apresentaram os maiores valores de redução de dureza (Capítulo 1).

Em se tratando da mensuração da rugosidade de superfície e perda de estrutura, como forma de mensuração da resistência à degradação de materiais resinosos, a utilização da perfilometria com imagem tridimensional mostrou ser precisa (Capítulo 2). Estudos anteriores evidenciaram a precisão do método, enfatizando a qualidade das leituras e a praticidade de poder realizar várias análises, principalmente aquelas relacionadas com perdas estruturais (Söderholm et al., 2001; Garcia-Godoy et al., 2009; Mueller et al., 2011; Cao et al., 2013).

Diante dos resultados obtidos na presente tese, pode-se observar que os dados foram bastante pertinentes para o conhecimento do comportamento dos materiais experimentais nos diferentes ensaios laboratoriais realizados, sendo a maioria das formulações consideradas semelhantes ou melhores que o infiltrante comercial. Os achados contribuíram para melhor caracterização dos materiais formulados e permitiu desenvolver critérios para selecionar as misturas que podem ser investigadas mais minuciosamente em testes futuros. A mistura à base de TEGDMA e UDMA parece ser a mais promissora e a adição da CHX não influenciou a maioria das propriedades avaliadas (sorção/solublididade, módulo de elasticidade, resistência à flexão e desgaste mecânico por três corpos).

4. CONCLUSÃO

Assim, diante dos resultados obtidos, pode-se concluir que:

1. A base monomérica influenciou o desempenho dos materiais para os testes de sorção/solubilidade, taxa de redução de dureza, módulo de elasticidade e resistência à flexão. Quando os monômeros UDMA ou BisEMA foram adicionados ao TEGDMA, aumentaram o módulo de elasticidade e a resistência à flexão; e diminuíram a sorção e solubilidade dos materiais testados, entretanto, não interferiram na taxa de redução de dureza.

2. Diferentes concentrações de CHX não interferiram nas propriedades de sorção/solubilidade, módulo de elasticidade e resistência à flexão; com exceção do teste de taxa de redução de dureza em que a mistura TEGDMA/UDMA/0,1%CHX apresentou maior valor, mas não foi diferente do controle comercial;

3. A resistência à abrasão mecânica por três corpos também foi dependente da base monomérica das misturas experimentais, sendo que a mistura TEGDMA/UDMA/0,2%CHX apresentou os menores valores de rugosidade e perda estrutural.

4. Medidas da rugosidade superficial em 2D (Ra) podem ser associadas às medidas da rugosidade superficial em 3D (Sa).

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* De acordo com as normas da UNICAMP/FOP, baseadas na pradronização do International Committee of Medical Journal Editors – Vancouver Group. Abreviatura dos periódicos em conformidade com o PubMed.

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APÊNDICES

Apêndice 1 – Figuras correspondentes aos procedimentos realizados no Capítulo 1.

Figura 1 – Molde metálico utilizado para obtenção do molde em polivinil silxano. A imagem mostra o molde utilizado para confecção dos espécimes cilíndricos.

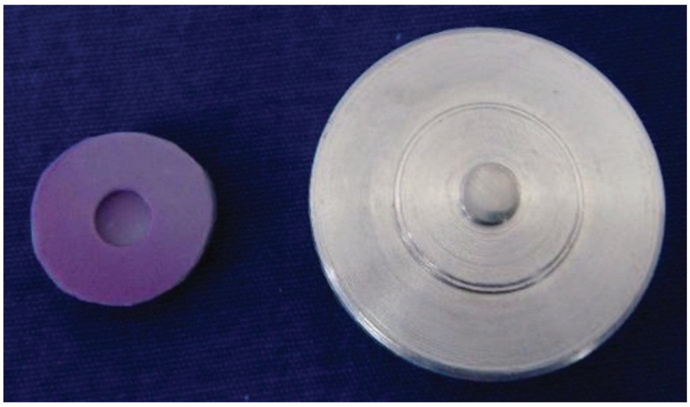


Figura 2 – Molde sendo preenchido pela mistura resinosa experimental.

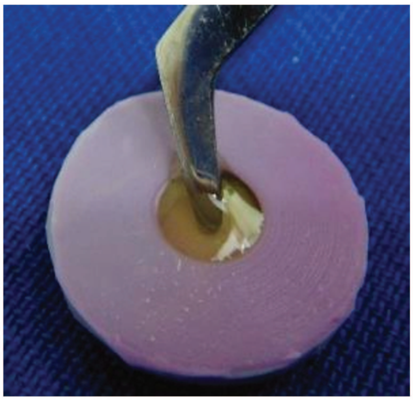


Figura 3 – Molde preenchido com a mistura resinosa coberto com tira de fita de poliéster e sobre esta uma lâmina de vidro.

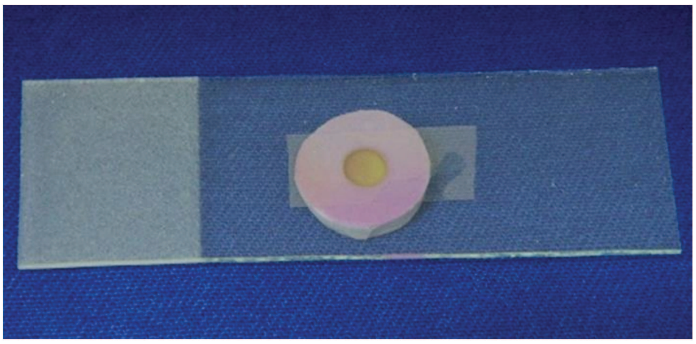


Figura 4 – Fotoativação por 60 s.

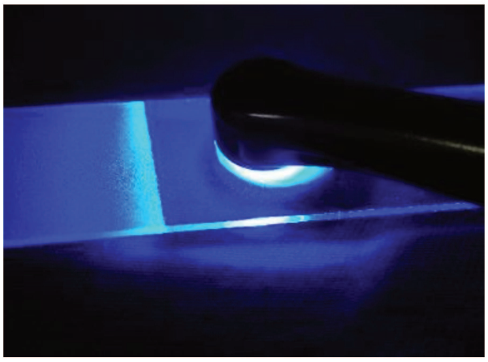


Figura 5 – Espécimes cilíndricos utilizados para os testes de sorção/solubilidade e densidade de ligações cruzadas.

Figura 6 – Espécimes em forma de barra utilizados para os testes de módulo de elasticidade e resistência à flexão.

Figura 7 – A)Teste de sorção/solubilidade - espécimes em processo de secagem por meio de dessecador à vácuo com sílica gel armazenados separadamente em Eppendorfs. B) Detalhe mostrando Eppendorfs abertos).

Figura 8 – Espécimes armazenados em água deionizada em tubos Falcon durante o teste de sorção/solubilidade.

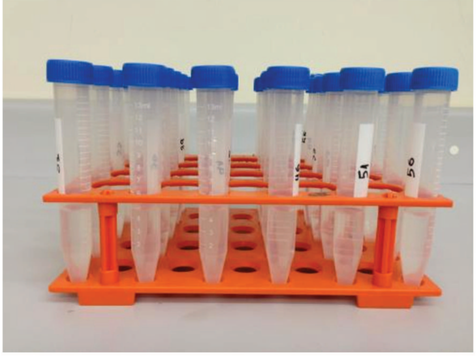


Figura 9 – Mensuração do diâmetro e altura dos espécimes cilindros para cálculo do volume do cilindro no teste de sorção/solubilidade.

Figura 10 – Microdurômetro Future Tech FM 100 (Future Tech Corp., Japão), utilizado para a avaliação da dureza Knoop para o teste de densidade de ligações cruzadas.

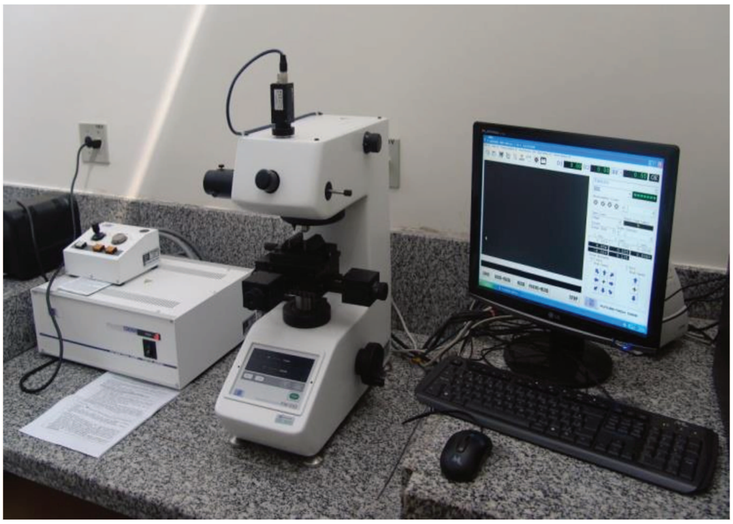


Figura 11 – Ponta Knoop realizando endentação na superfície do espécime.

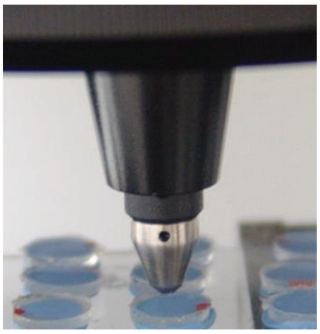


Figura 12 – Imagem da endentação gerada pelo software do aparelho.

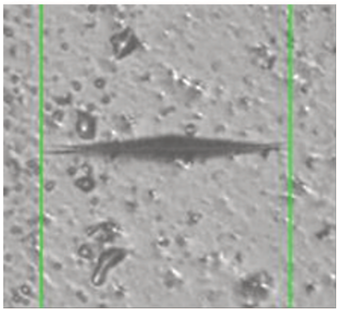


Figura 13 – Máquina Universal INSTRON, modelo 4111 (Instron Corp., EUA), utilizada para os testes de módulo de elasticidade/resistência à flexão.

Apêndice 2 – Figuras correspondentes aos procedimentos realizados no Capítulo 2.

Figura 1 – Molde metálico e molde em polivinil siloxano para confecção de espécimes em forma de barra utilizadas no teste de abrasão por três corpos.

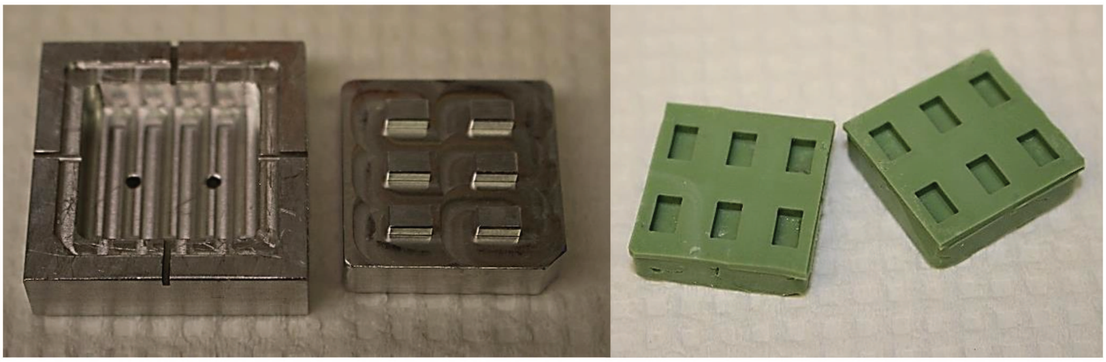


Figura 2 – Molde sendo preenchido com mistura experimental.

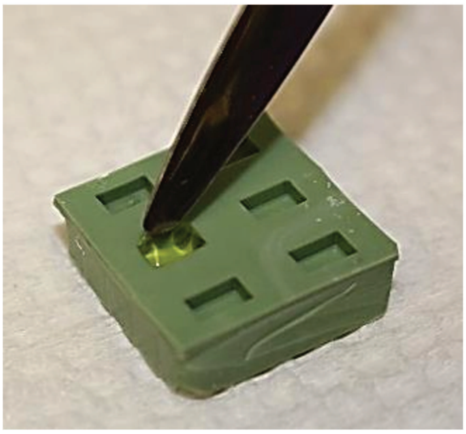


Figura 3 – Molde com mistura experimental coberta por lâmina de vidro.

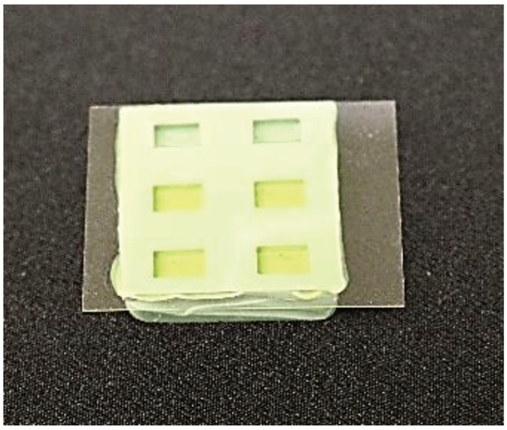


Figura 4 – Fotoativação por 60 s.

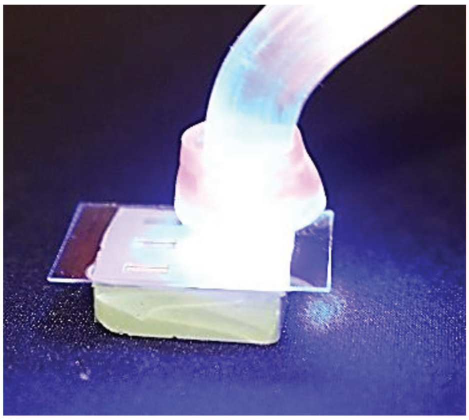


Figura 5 – Espécimes prontos.

Figura 6 – Molde em teflon usado para embutir espécimes.

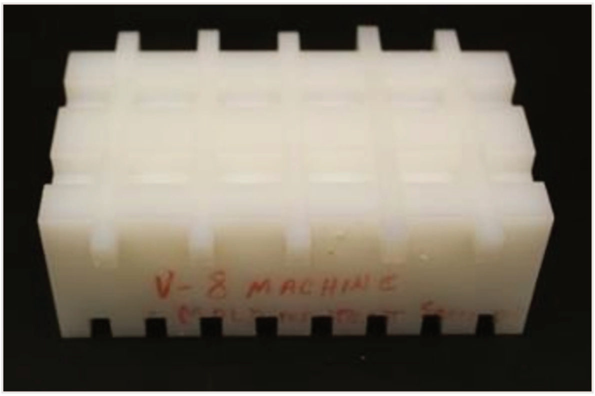


Figura 7 – Espécimes posicionados no molde de teflon.

Figura 8 – Preenchimento dos nichos retangulares do molte e teflon com resina acrílica para base de dentadura.

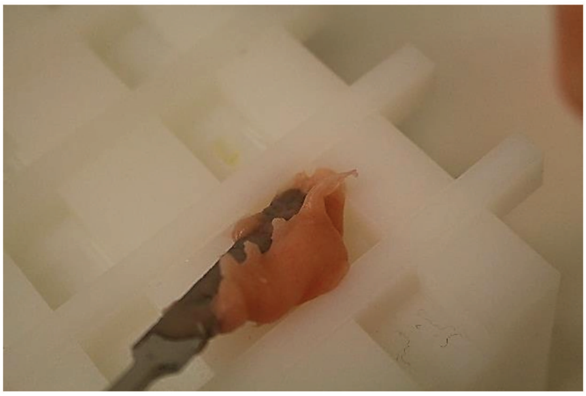


Figura 9 – Resina acrílica polimerizada.

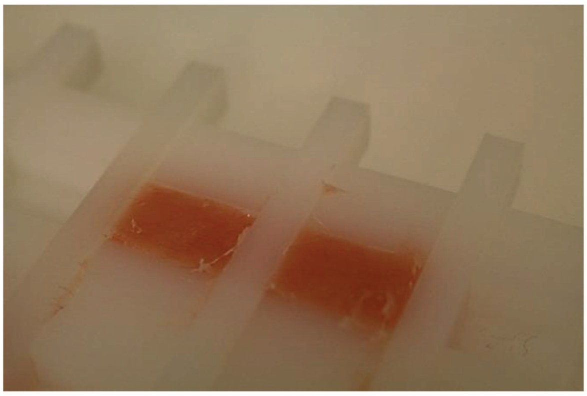


Figura 10 – Removendo os espécimes embutidos do molde de teflon. .

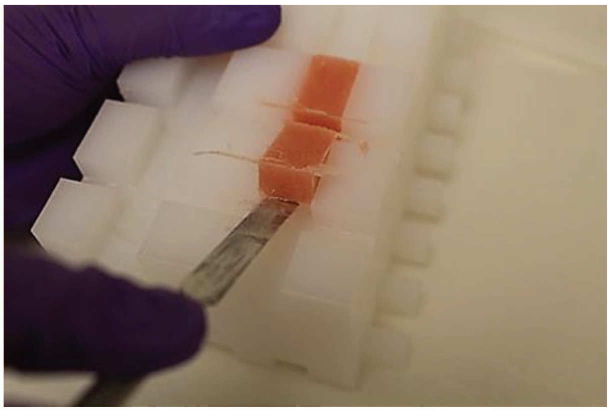


Figura 11 – Espécimes embutidos prontos.

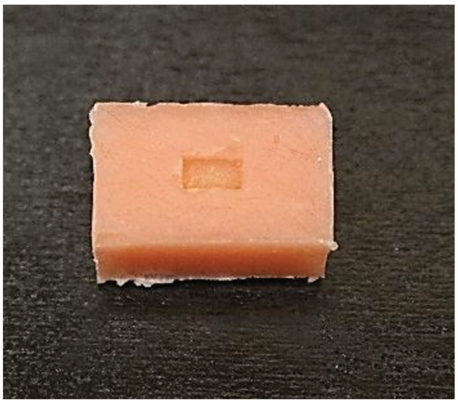


Figura 12 – Posicionando espécimes em dispositivo da politriz para polimento das superfícies superior e inferior dos blocos de resina acrílica.

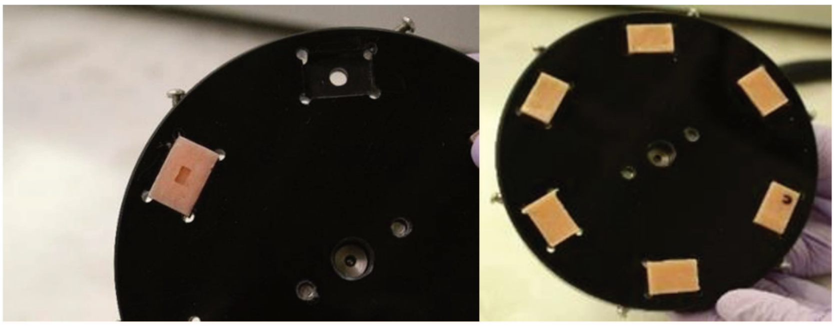


Figura 13 – A) Politriz AutoMet 250 (Buehler, EUA). B)Detalhe mostra espécimes durante processo de polimento.

Figura 14 – Sequência de discos para polimento e pasta diamantada.

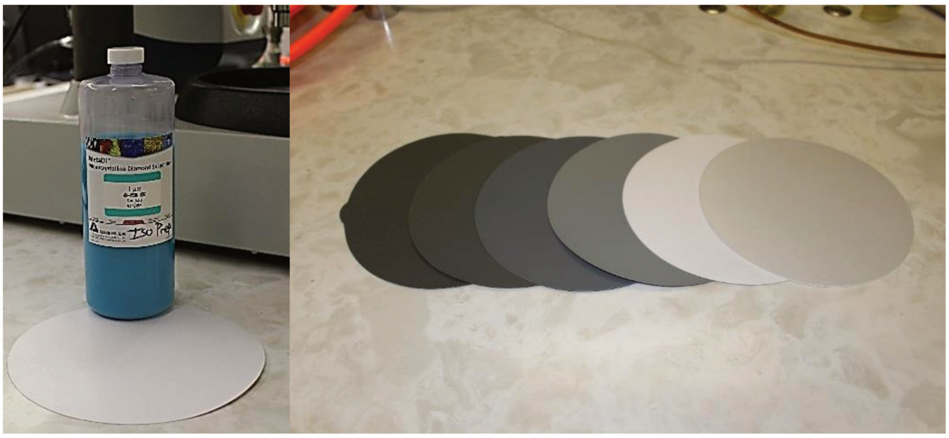


Figura 15 – Conferindo paralelismo das superfícies dos blocos de resina acrílica com paquímetro.

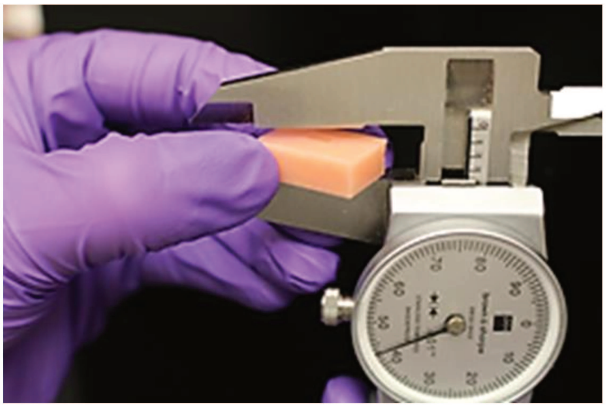


Figura 16 – Perfilômetro Taylor Hobson CCI (Ametek, EUA). Detalhe acima mostra escaneamento dos espécimes sem contato com a superfície.

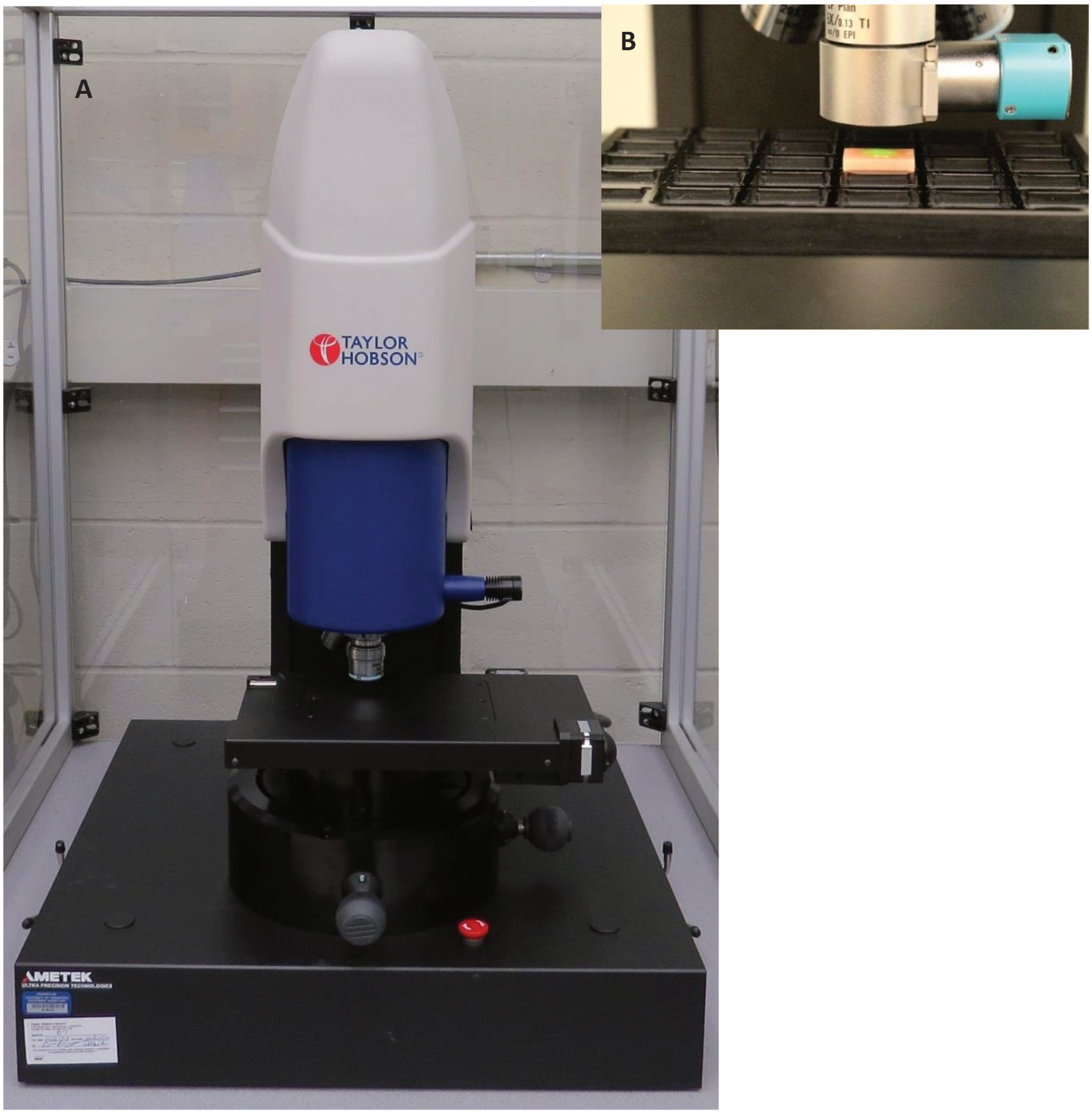


Figura 17 – Delimitação da área a ser escovada com adesivo (Argon Inc., CA, EUA).

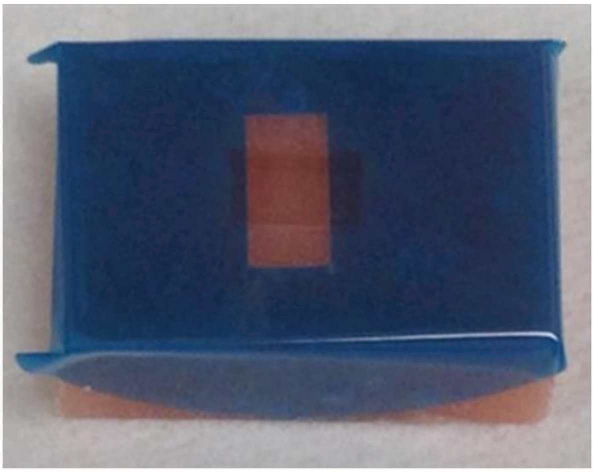


Figura 18 – Posicionamento das certas da escova sobre a área a ser escovada.

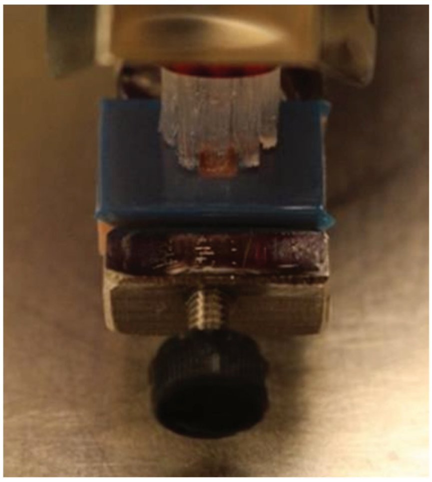


Figura 19 – Padronização de força para escovação por meio de dinamômetro.

Figura 20 – Pesagem da água deionizada e dentifrício na proporção 1:1 para preparo do *slurry*.

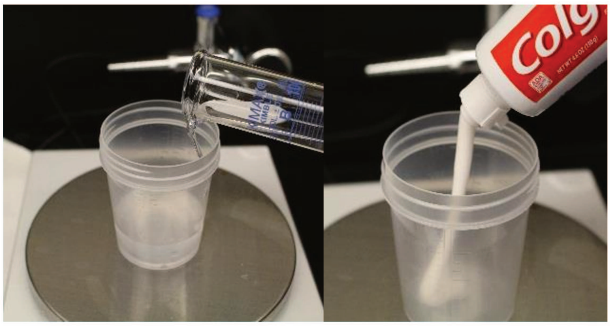


Figura 21 – Mistura da água e dentifrício e aspecto do *slurry* pronto.

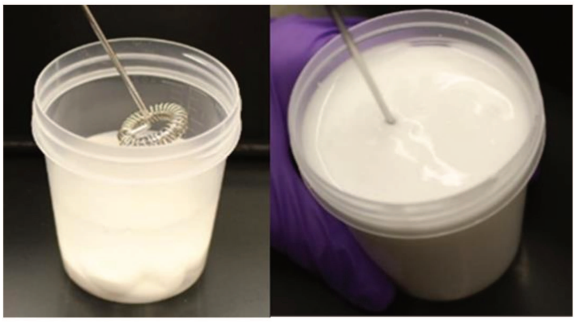


Figura 22 – Máquina de escovação modelo V8 durante os ciclos de escovação.

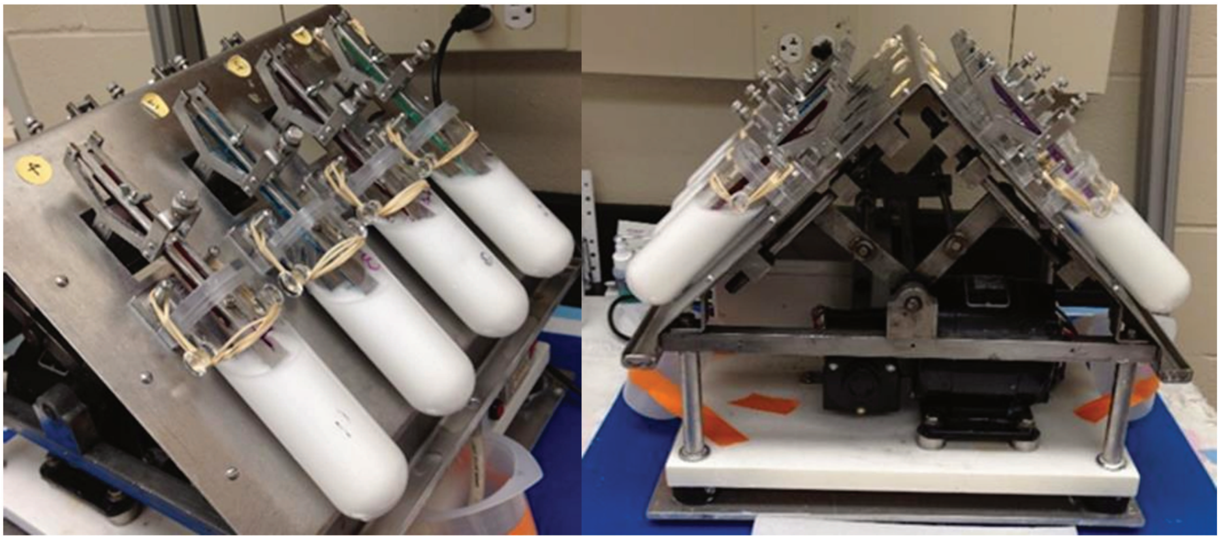


Figura 23 – Aspecto final da área escovada.

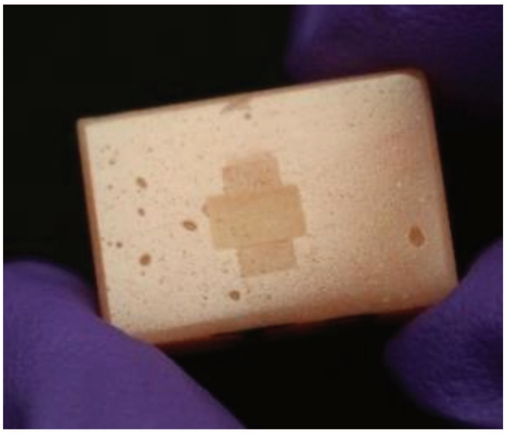
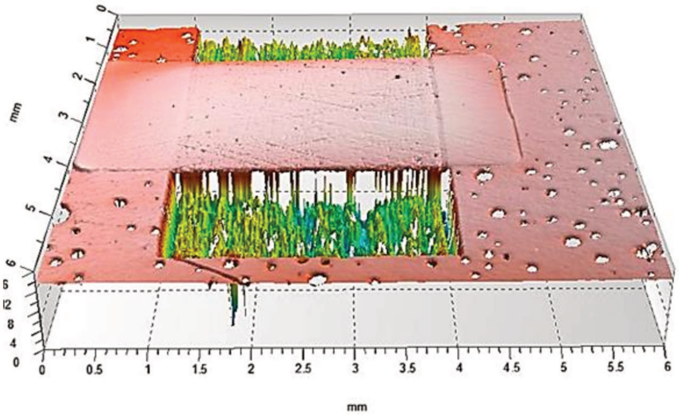


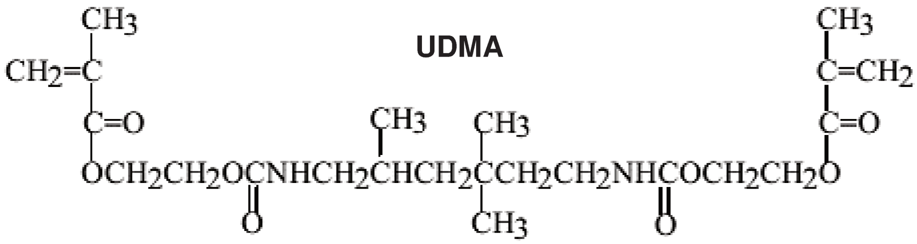
Figura 24 – Exemplo de imagem 3D da área escovada obtida pelo perfilômetro.



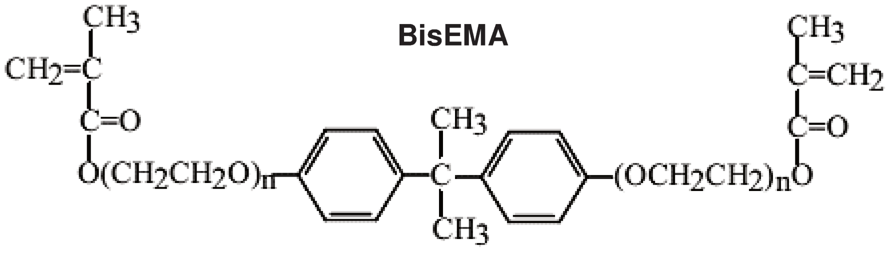
ANEXOS

Anexo 1 – Figuras correspondentes à estrutura química dos monômeros TEGDMA, UDMA e BisEMA (componentes das misturas resinosas experimentais) e do monômero BisGMA[4].

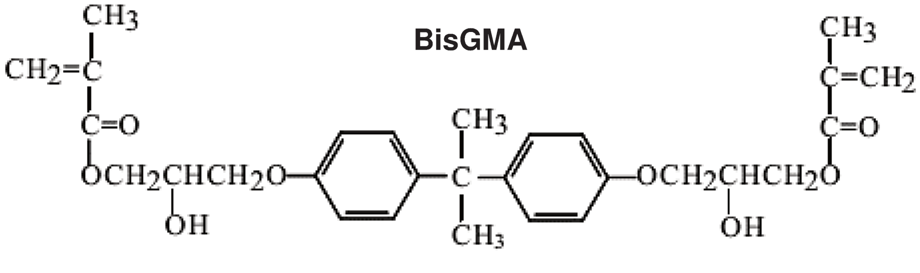
TEGDMA



UDMA



BisEMA



BisGMA

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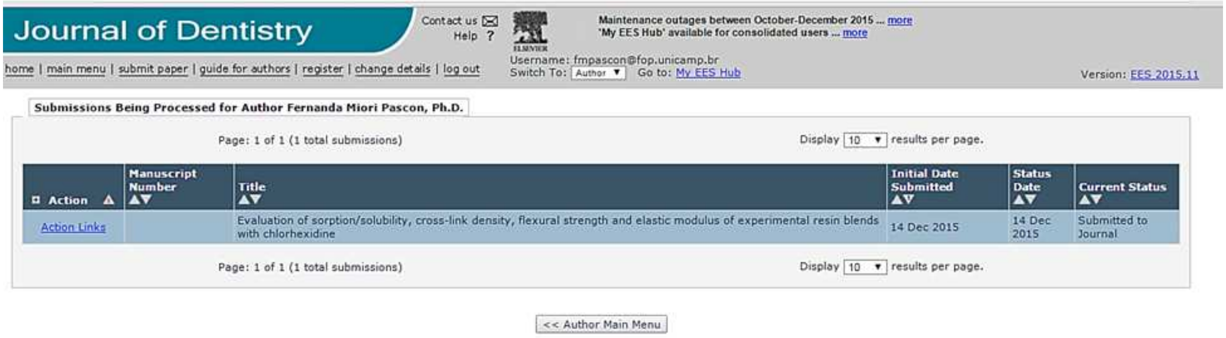
[4] Fonte das figuras: Sideridou I, Tserki V, Papanastasiou G. Effect of chemical structure on degree of conversion in light-cured dimethacrylate-based resins. Biomaterials. 2002; 23(8): 1819-29.

Anexo 2 – Comprovante de submissão de artigo científico (Capítulo 1):



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Action	Manuscript Number	Title	Initial Date Submitted	Status Date	Current Status
Action Links		Evaluation of sorption/solubility, cross-link density, flexural strength and elastic modulus of experimental resin blends with chlorhexidine	14 Dec 2015	14 Dec 2015	Submitted to Journal

Page: 1 of 1 (1 total submissions)

Dear Prof. Pascon,

Your submission entitled "Evaluation of sorption/solubility, cross-link density, flexural strength and elastic modulus of experimental resin blends with chlorhexidine" has been received by the Journal of Dentistry.

You will be able to check on the progress of your paper by logging on to Elsevier Editorial System as an author. The URL is http://ees.elsevier.com/jjod/.

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