UNIVERSIDADE DE SAO PAULO
FACULDADE DE ODONTOLOGIA DE BAURU

FABIO ANTONIO PIOLA RIZZANTE

Mechanical and physical properties assessment of Bulk Fill resin
composites

Avaliação das propriedades físico-mecânicas de resinas compostas do
tipo Bulk Fill

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Dedico essa conquista à Deus e à minha família, sem os quais não
teria chego até aqui.

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“No seu último dia de vida, a pessoa que você se tornou irá
encontrar a pessoa que você poderia ter se tornado”

– adaptado (autor desconhecido)

“Independente do que o amanhã me traga, eu estarei lá, de
braços e olhos abertos”

– tradução livre de um trecho da música Drive, da banda Incubus

ABSTRACT

Mechanical and physical properties assessment of Bulk Fill resin composites

The achievement of predictable and long lasting adhesive restorations in posterior teeth have always been a major objective of studies in the context of materials and techniques development. The use of bulk fill composites could provide better outcomes, but it is important to assess their physico-mechanical properties, responsible for their clinical behavior. The purpose of the present study was to assess the mechanical and physical properties of bulk fill composites. The composites were divided into 2 groups according to their viscosity. For low viscosity composites, the present study assessed: Surefill SDR flow (SDR), X-tra Base (XB), Filtek Bulk Fill Flowable (FBF), and Filtek Z350XT Flow (Z3F-control); and for high viscosity composites: Tetric Evo Ceram Bulk Fill (TBF), X-tra Fil (XF), Filtek Bulk Fill (FBP), Admira Fusion x-tra (ADM) and Filtek Z350 XT (Z3XT-control). Composites were assessed through shrinkage stress test (using 12 and 24mm³ of composite in a custom device adapted in an Universal Testing Machine); volumetric shrinkage (using 64mm³ of composite placed on a Teflon mold and scanned in a micro computed tomography/ μ CT); Young's modulus (through a 3-point bending test device adapted in an Universal Testing Machine); microhardness and depth of cure tests (using longitudinal Knoop microhardness). All data was evaluated regarding their homogeneity using Shapiro-Wilk test. For polymerization stress, 3-way Variance Analysis (ANOVA) was used. Considering Volumetric Shrinkage, Young's Modulus, Microhardness and Depth of Cure, one-way ANOVA was used. All ANOVA tests were followed by Tukey's test and 5% was adopted as significance level. Shrinkage stress test with 12mm³ showed SDR, TBF and XF generating the lowest stress after 300s, followed by other high viscosity composites (ADM, FBF, XB and FBP/Z3XT). The regular low viscosity composite (Z3F) generated the highest stress for all assessed times. Considering the same test, with 24mm³, after 300s, SDR, FBP and ADM generated similar stress, followed by TBF and XF. Low viscosity bulk fill composites generated lower stress than Z3F. Considering Young's modulus, low viscosity composites (SDR, FBF, XB and Z3F) showed the lowest values, followed by ADM and TBF. The other high viscosity composites (Z3XT, FBP and XF) showed the highest values. For microhardness test, all low viscosity composites showed lower values (FBF being the

lowest). For high viscosity composites, Z3XT showed the highest values, followed by XF, FBP/TBF and ADM. Assessing depth of cure, regular composites showed lower values when compared with bulk fill composites. All bulk fill composites showed adequate depth of cure over 4.5mm (microhardness $\geq 80\%$ of initial reading). SDR and XB showed the highest depth of cure. All high viscosity bulk fill composites generated lower volumetric shrinkage than regular composites. All low viscosity

bulk fill composites showed similar volumetric shrinkage when compared to the regular composites (Z3F and Z3XT). Bulk fill composites show characteristics that allow their use in larger increments (i.e. volumetric shrinkage and polymerization stress similar or lower when compared with regular composites). Nonetheless, the mechanical properties of bulk fill composites were widely variable, being important to individually assess each material previously to its clinical application.

Keywords: Bulk Fill Resin Composites; Restoration; Shrinkage stress, Biomechanics, Operative Dentistry, Micro-computed tomography

RESUMO

A obtenção de restaurações adesivas previsíveis e duradouras em dentes posteriores sempre foi objetivo de estudos na área de desenvolvimento de materiais e técnicas. O uso de resinas compostas do tipo bulk fill pode possibilitar melhores resultados, porém é importante o estudo de suas propriedades físico-mecânicas, responsáveis por seus comportamentos clínicos. O objetivo do presente estudo foi o de avaliar as propriedades físicas e mecânicas das resinas bulk fill. As resinas foram divididas em 2 grupos de acordo com sua viscosidade. Para resinas de baixa viscosidade, o presente estudo avaliou: Surefill SDR flow (SDR), X-tra Base (XB), Filtek Bulk Fill Flowable (FBF) e Filtek Z350XT Flow (Z3F-controle); e, para alta viscosidade: Tetric Evo Ceram Bulk Fill (TBF), X-tra Fil (XF), Filtek Bulk Fill (FBP), Admira Fusion x-tra (ADM) e Filtek Z350 XT (Z3XT-controle). As resinas foram avaliadas em relação à tensão de polimerização (utilizando 12 e 24mm³ de resina adaptadas em um dispositivo adaptado a uma máquina de testes universal); contração volumétrica (utilizando 64mm³ de resina composta inserida em um molde de Teflon e escaneada em um micro-tomógrafo/ μ CT), módulo de Young (através de um dispositivo de flexão em 3 pontos adaptado a uma máquina de testes universal), microdureza e profundidade de polimerização (utilizando microdureza Knoop). Todos os resultados foram avaliados em relação à homogeneidade utilizando o teste de Shapiro-Wilk. Para avaliação da tensão de polimerização, foi empregada a Análise de Variância (ANOVA) a 3 critérios. Para as análises de contração volumétrica, Módulo de Young, microdureza e profundidade de polimerização, ANOVA a um critério foi empregada. Todas as Análises de Variância foram seguidas pelo teste de Tukey e 5% foi adotado como nível de significância. A tensão de polimerização com 12mm³ demonstrou que SDR, TBF e XF geraram valores significativamente mais baixos após 300s, seguidas por outras resinas de alta viscosidade (ADM, FBF, XB e FBP/Z3XT). A resina convencional de baixa viscosidade (Z3F) gerou valores de tensão significativamente mais elevados para todos os tempos avaliados. Considerando o mesmo teste, com 24mm³, após 300s, SDR, FBP e ADM geraram valores estatisticamente inferiores, seguidas por TBF e XF. As resinas bulk fill de baixa viscosidade geraram menor tensão de polimerização que a Z3F. Considerando o módulo de Young,

resinas de baixa viscosidade (SDR, FBF, XB e Z3F) apresentaram valores significativamente inferiores, seguidas por ADM e TBF. As outras resinas de alta viscosidade (Z3XT, FBP e XF) apresentaram valores significativamente mais elevados. Para o teste de microdureza, todas as resinas de baixa viscosidade apresentaram valores inferiores (FBF apresentou o menor). Para as resinas de alta viscosidade, Z3XT apresentou os valores mais elevados, seguida por XF, FBP/TBF e ADM. Para profundidade de polimerização, resinas compostas convencionais apresentaram valores significativamente mais baixos quando comparadas com resinas bulk fill. Todas as resinas bulk fill apresentaram profundidade de polimerização adequada até pelo menos 4,5mm (microdureza $\geq 80\%$ da leitura inicial/superfície). SDR e XB apresentaram os valores mais altos de profundidade de polimerização. Todas as resinas bulk fill de alta viscosidade geraram menor contração volumétrica que resinas compostas convencionais. Todas as resinas bulk fill de baixa viscosidade apresentaram contração volumétrica similar às resinas convencionais (Z3F e Z3XT). Resinas compostas bulk fill apresentaram características que possibilitam sua indicação para serem empregadas em grandes incrementos (contração volumétrica e tensão de polimerização similar ou inferiores às resinas convencionais, além de maior profundidade de polimerização). No entanto, as propriedades mecânicas variaram grandemente entre as resinas estudadas sendo importante uma avaliação individual de cada material previamente ao seu uso clínico.

Palavras-chave: Resina Composta Bulk Fill, Stress de polimerização, Biomecânica, Dentística Operatória, Micro Tomografia Computadorizada

List of Legends and Acronyms

ADM	Admira Fusion x-tra
FBP	Filtek Bulk Fill Posterior
FBF	Filtek Bulk Fill Flowable
SDR	Surefil SDR flow
TBF	Tetric Evo Ceram Bulk Fill
XB	X-tra Base
XF	X-tra Fil
Z3F	Filtek Z350 flow
Z3XT	Filtek Z350XT
Bis-GMA	Bisphenol A glycidyl methacrylate
Bis-EMA	Ethoxylated bisphenol A dimethacrylate
TEGDMA	Triethylene glycol dimethacrylate
EBPDMA	Ethoxylated bisphenol A dimethacrylate
UDMA	Urethane dimethacrylate

SUMMARY

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1. INTRODUCTION

1. INTRODUCTION

Adhesive restorations in posterior teeth, as treatment of dental caries or replacement of metallic or unsatisfactory restorations consist of a basic procedure in daily clinical practice (MANHART et al., 2004). However, there are still many doubts about the restorative protocol due to the wide variety of available (and in development) materials. In addition, the wide variety of restorative techniques, regarding the cavity preparation and adhesive systems, can also generate questions for professionals (DIETSCHI et al., 2013; PECIE et al., 2013; TEIXEIRA et al., 2016).

Adhesion provided a major breakthrough in cosmetic dentistry. Due to composites' properties evolution, they started to be used in areas subjected to greater masticatory forces such as posterior restorations. Despite its excellent properties, clinically they are still more prone to failures in posterior areas due to wear, medium to long term deterioration of the adhesive interface, technical sensitivity, polymerization shrinkage and inadequate polymerization, especially in class II with cervical margins located in dentin or cementum (ARAUJO FDE et al., 2006; DHINGRA et al., 2014).

In general, small cavities are restored with direct restorative materials while extensive cavities can be restored with indirect procedures such as ceramic restorations. However, the reference to "intermediate" restorations is limited due to the lack of long term clinical reports (DIETSCHI et al., 2013).

For direct restorations in posterior teeth, the literature reports 5 main factors that can lead to failure. Besides improper handling of restorative materials, marginal defects and secondary caries (failure in the adhesive interface), restoration fracture and excessive wear (failure in the material) are also reported (DEMARCO et al., 2001; MANHART et al., 2004).

Defects in the adhesive interface are generated by the characteristics of resinous materials. During polymerization process, composites generate shrinkage stress, accompanied by a variable volumetric shrinkage (ILIE, HICKEL, 2011; ISHIKIRIAMA et al., 2013; KIM, PARK, 2014; MOORTHY et al., 2012). Polymerization stress at the adhesive interface is dependent on the organic/inorganic composition of resins, as well as on the viscoelastic properties of materials (BRAGA et al., 2003). Clinically, the generated tension is also dependent on the cavity configuration (C-Factor), given by the proportion between adhered surfaces area and free surfaces area. The increase in C-Factor is associated with a reduction in bond strength and loss of marginal integrity. The presence of micro gaps at the interface may lead to microleakage of saliva and bacteria, adhesive interface

degradation, secondary caries and pulpal changes, resulting in clinical failure of the restoration (DEJAK, MLOTKOWSKI, 2015; DUARTE et al., 2007; FERRACANE, HILTON, 2016; HASHIMOTO et al., 2003), however, if the bond strength is greater than the generated tension, enamel cracks and/or cusps deflection can occur (MOORTHY et al., 2012).

The stress at the adhesive interface is more relevant when the restoration's margins are defined in dentin or cementum since, due to the high organic content when compared to enamel, the adhesive properties are jeopardized (DHINGRA et al., 2014).

Many operatory techniques associated with different restorative materials have been studied in order to minimize marginal integrity failures. Considering the different restorative techniques, incremental insertion technique is recommended to ensure a better marginal integrity due to prevention of stresses development at the adhesive interface (DIETSCHI et al., 2002; ILIE, HICKEL, 2011; KRAMER et al., 2011; MOORTHY et al., 2012; VAN DIJKEN, PALLESEN, 2011). However, despite the incremental technique advantages, ensuring a better polymerization and stress distribution, it's more laborious and time costing, creating a risk of air bubbles trapping between the resin layers, which can result in lower mechanical properties (CAMPOS et al., 2014).

Despite constant advances in adhesive systems, changes in resin composites are not so frequent, thus, the use of nanotechnology enabled the development of composites that can be used for both anterior and posterior restorations. Such technology allowed an increase in the inorganic filler content, which could be related with lower polymerization shrinkage and better physico-mechanical and optical properties (CAMPOS et al., 2014; MITRA et al., 2003; ROCHA GOMES TORRES et al., 2014).

With the popularization of bulk fill composites, based on the already discussed advantages, some aspects should be analyzed: the polymerization shrinkage, volumetric shrinkage and Young's modulus (associated with the tooth/restoration interface stress) and the microhardness/depth of cure that can be compromised due to use of larger increments.

Besides the increase of filler content in composites, another attempt to reduce the polymerization stress was the substitution of Bis-GMA for other monomers like silorane. This modification resulted in a low shrinkage resin but with inadequate mechanical properties and conflicting results regarding their shrinkage stress (ISHIKIRIAMA et al., 2014; PORTO et al., 2013; BOARO et al., 2010). More recently, bulk fill resin composites were developed, with different rheological properties, allowing their use as base or restorative materials (EL-SAFTY et al., 2012; GARCIA et al., 2014; ILIE, HICKEL, 2011; MOORTHY et al., 2012). Another important property of this class of composites relies on the possibility of using larger

increments without compromising the degree of conversion (up to 4mm according to manufacturers). This became possible due to a better control of the polymerization process and use of nanotechnology.

In order to allow larger increments insertion, the molecular basis of these composites was changed by substitution of the Bis-GMA for monomers with higher molecular weight such as Bis-EMA, TEGDMA, EBPDMA and UDMA monomers, as well as adopting stress relievers (BURGESS, CAKIR, 2010; CZASCH, ILIE, 2013; ILIE, HICKEL, 2011; JANG et al., 2015; KIM et al., 2015). One may question about the organic and inorganic filling of these composites, since “conventional” ones sometimes shares the same composition but, unfortunately, the proportion between the different monomers is not reported by the manufacturers (BURGESS, CAKIR, 2010; EL-DAMANHOURY, PLATT, 2014; ILIE, HICKEL, 2011; KIM et al., 2015). Nevertheless, the related chemical modifications resulted in composites with slower polymerization rate and reduced polymerization stress (BURGESS, CAKIR, 2010; EL-DAMANHOURY, PLATT, 2014; ILIE, HICKEL, 2011).

Clinically, all those benefits reflects in an easier cavity restoration, with fewer number of increments and shorter handling time (GARCIA et al., 2014). However, there is still insufficient information about the biomechanical behavior of these materials as well as their impact on the adaptation to the dental structures, cusp deflection, among other factors (EL-DAMANHOURY, PLATT, 2014).

This *in vitro* study had as goals to address the physical and mechanical properties of bulk fill composite resins through the assessment of polymerization shrinkage stress, volumetric shrinkage, Young's modulus, microhardness and depth of cure. For these purposes, two articles are presented. The first article evaluated the polymerization shrinkage stress of different bulk fill resin composites and their Young's modulus; and the second article evaluated the polymerization shrinkage, Knoop microhardness and depth of cure of the different composites.

2. ARTICLES

2. ARTICLES

- **Article 1** - Mechanical properties of Bulk Fill resin composites
- **Article 2** – Polymerization shrinkage, microhardness and depth of cure of bulk fill resin composites

2.1. ARTICLE 1

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Mechanical properties of Bulk Fill resin composites

Bulk Fill composites mechanical properties

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ABSTRACT

Objectives. To evaluate the polymerization shrinkage stress of different bulk fill resin composites and their Young's modulus, compared with regular composites

Materials and Methods. Fourteen specimens were made for each of the nine different resin composites (seven with 12mm³ and seven with 24mm³). Materials were divided into 2 groups based on their viscosity - low viscosity: Surefill SDR flow (SDR), X-tra Base (XB), Filtek Bulk Fill Flowable (FBF), Filtek Z350XT Flow (Z3F- control); and high viscosity: Tetric Evo Ceram Bulk Fill (TBF), X-tra Fil (XF), Filtek Bulk Fill (FBP), Admira Fusion x-tra (ADM) and Filtek Z350 XT (Z3XT- control). The linear shrinkage stress was addressed during 300s with the aid of a linear shrinkage device adapted in a Universal Testing Machine. In addition, seven specimens (2x2x25mm) for each group were made and the Young's modulus was evaluated with a 3-point bending device adapted in the Universal Testing Machine with crosshead speed of 0.5mm/min and 50KgF loading cell.

Results. For 12mm³, three-way ANOVA followed by Tukey's test showed that SDR, TBF and XF generated the lowest stress after 300s, followed by ADM, FBF, XB, and FBP, which were similar to Z3XT. Z3F generated the highest stress values for all time points. Considering 24mm³, after 300s, SDR, FBF and ADM generated the lowest stress, followed by TBF and XF. For Young's modulus, one-way ANOVA showed that SDR, FBF, XB, and Z3F presented the lowest values, followed by ADM and TBF. Z3XT, FBP, and XF presented the highest elastic modulus among the evaluated composites.

Conclusions. Bulk fill resin composites present equal to lower shrinkage stress generation when compared with conventional composites, mainly with bigger increments. Bulk fill composites show a wide range of elastic modulus values, but usually similar to "regular" composites.

Clinical Significance. These findings show the rationale for using bulk fill composites since they generate similar to lower shrinkage stress when compared with "regular" composites. The heterogeneous results show also the importance of consider each composite individually.

Keywords: Bulk Fill Resin Composites; Restoration; Shrinkage stress

1. INTRODUCTION

Despite advances in adhesive dentistry, composite resins still tend to fail in extensive posterior restorations due to wear, medium to long term adhesive interface deterioration, technical sensitivity, polymerization shrinkage and inadequate polymerization, especially in class II with cervical margins located in dentin or cementum [1,2]. Defects in the adhesive interface are generated by the characteristics of resinous materials. During the polymerization process, composites suffers shrinkage (polymerization shrinkage) that is dependent on the material composition and volume [3,4]. This shrinkage can generate tensions at the adhesive interface between the restorative material and the dental structures, which may cause the formation of micro gaps and, therefore, allow the microleakage of saliva and bacteria. This effect can lead to the adhesive interface degradation, secondary caries and pulpal changes, resulting in clinical failure of the restoration [5].

Incremental insertion technique is recommended to ensure a better marginal integrity due to reduction of polymerization shrinkage stresses development [6-8]. However, despite the incremental technique advantages, ensuring a better polymerization and stress distribution, it is more laborious, technical sensitive and time costing [9].

Bulk fill resin composites are advised to be used in larger increments without compromising the degree of conversion (up to 4mm according to some manufacturers). A major concern with the polymerization of large increments relies on the polymerization shrinkage and in the tensions generated at the tooth/restoration interface [10,11]. Promising results have been reported with these materials, mainly due to the lower polymerization shrinkage [12], which also depends on the composite organic/inorganic matrix composition and properties such as viscosity and elastic modulus.

Although several materials with varying viscosities and handling characteristics are commonly classified as bulk fill resin composites, their properties may change considerably, especially due to modifications in the organic matrix, with incorporation of monomers with higher molecular weight, as well as changes in filler content and incorporation of stress relievers [10,13-16].

Composites can be subdivided according to its consistency in low and high viscosity. Higher shrinkage stress for flowable composites are expected since generally they have a higher organic content when compared with microhybrid and nanoparticulated ones, which can result in greater polymerization shrinkage and lower mechanical properties [15,17]. Similarly, a lower Young's modulus may allow stress dissipation during polymerization

process, thus reducing the stress with bigger increments [18,19]. This said, viscoelastic behavior (and its development during the polymerization process), and volumetric shrinkage play major roles during the polymerization stress generation, demonstrating the importance for stress development among composites with different viscosities [18,19]. Thus, the aim of the present study was to access the polymerization shrinkage stress and the elastic modulus of different bulk fill resin composites, compared with regular composites.

2. MATERIALS AND METHODS

Nine different resin composites were evaluated in this study (Table 1) having as response variables: linear shrinkage stress (considering increment's volume in 2 levels: 12mm³ and 24mm³), and Young's modulus.

Table 1 – Different groups with respective composition and manufacturer's

Group	Restorative material	Organic matrix composition	Filler weight (%)
ADM	Admira Fusion x-tra, VOCO, Cuxhaven, Germany	Ormocer resin	84%
FBP	Filtek Bulk Fill Posterior, 3M ESPE, St Paul, MN, USA	AUDMA, UDMA and 1, 12-dodecane-DMA	76.5%
FBF	Filtek Bulk Fill Flowable, 3M ESPE, St Paul, MN, USA	UDMA, BISGMA, Bis-EMA, Procrylat resin	64.5%
SDR	Surefil SDR flow, Dentsply Sirona, York, PA, USA	Modified UDMA, EBPADMA, TEGDMA	68%
TBF	Tetric Evo Ceram Bulk Fill, Ivoclar Vivadent, Schaan, Liechtenstein	Bis-GMA, UDMA	78%
XB	X-tra Base, VOCO, Cuxhaven, Germany	UDMA, Bis-EMA	75%
XF	X-tra Fil, VOCO, Cuxhaven, Germany	Bis-GMA, UDMA, TEGDMA	86%
Z3F	Filtek Z350 flow, 3M ESPE, St Paul, MN, USA	Bis-GMA, Bis-EMA, TEGDMA	65%
Z3XT	Filtek Z350XT, 3M ESPE, St Paul, MN, USA	Bis-GMA, Bis-EMA, UDMA, TEGDMA	82%

2.1. Linear shrinkage stress

Tensilemetry was used to evaluate the linear polymerization stress of the composites [4,20]. For this test, seven 12mm³ and seven 24mm³ specimens of each resin were used.

The restorative materials were inserted between two metallic bases, with 6 x 2mm as surface dimensions (Figure 1). These bases were previously sandblasted (surfaces in contact with composites) with aluminum oxide, avoiding the necessity of adhesive system application.

Figure 1- A- Schematics of the testing device; B- Universal testing machine (INSTRON) and metallic bases with specimen in position.

The metallic bases were adapted on a universal testing machine (Instron, model 3342, Norwood, MA, USA) with specific devices (Figure 1). The upper base was adapted on the testing machine articulated arm through a 50Kgf load cell. The lower base was fixed to the machine through a BENCOR multi testing device.

To standardize the material volume, the composite was inserted between the bases, with 1mm between them, resulting in a 12mm^3 constant volume and a 1.5 C-factor. The same test was repeated with 2mm between the bases, resulting in 24mm^3 and 0.75 C-factor. Light curing was performed during 20s, over the 6mm surface, with a $1550\text{mW}/\text{cm}^2$ irradiance ($31\text{J}/\text{cm}^2$ radiant exposure) LED light curing unit, LED Blue Star 3 (Microdont, São Paulo, SP, Brazil), assessed with a radiometer. The polymerization shrinkage induced tensions were analyzed by a specific software through the 50Kgf load cell deformation, since the articulated arm was set to be fixed during the tests. The traction force, induced by composite's shrinkage, allowed a real-time evaluation of the forces generated during the polymerization, assessed during 300s, starting from light curing. These data were registered as force (Newtons) X time (seconds) graphs and converted to MPa dividing the force results by the area of specimens' transversal section (12mm^2).

2.2. Young's modulus tests

For Young's modulus test, a three-point bending test was used. Seven specimens were made for each resin through insertion of the composite into a metallic matrix ($2 \times 2 \times 25\text{mm} = 100\text{mm}^3$ – ISO 4049) isolated with a specific product (Gel tripla ação, KG Sorensen, Cotia, SP, Brazil).

The specimen's dimensions were standardized by positioning polyester strips (Kdent, Quimidrol, Joinville, SC, Brazil) on both upper and lower surfaces before light curing. The polymerization was performed on both upper and bottom surfaces, in three points (left edge, right edge and center), during 40s for each one, according to ISO 4049 recommendations, in a total of 240s and $372\text{J}/\text{cm}^2$.

Specimens were removed from the matrix and stored in 37°C distilled water during 24 hours, in absence of light. After this, excesses were removed with a silicon carbide paper disk, with 1200grit (Buehler Ltd., Lake Bluff, IL, USA) adapted in a polisher. Specimens were adapted in a flexural test device, with 3 metal barrels. Two of these (separated by 20mm) were positioned on the specimen's bottom. The third cylinder was positioned on the central upper surface and was responsible for the flexural tension. The set was adapted in an universal testing machine and the force was applied through the upper articulated arm of the Instron, with $0.5\text{mm}/\text{min}$ crosshead speed, until specimen fracture (Figure 2). Based on the results, Young's modulus was determined, using the built-in software.

Figure 2 – Three-point bending device with sample.

2.3. STATISTICAL ANALYSIS

For all the statistical analysis, 5% was adopted as significance level ($p \leq 0.05$). All data was evaluated for results homogeneity using Shapiro-Wilk test. For polymerization

shrinkage stress, 3-way ANOVA was used (time, composites and volume). For Young's modulus assessment, one-way Anova was performed. All the above mentioned parametric tests were followed by Tukey's test.

In addition, a linear regression analysis was performed considering Young's modulus and filler content (Figures 4 and 5), as well as considering Young's modulus and shrinkage stress (Figures 6 and 7).

3. RESULTS

The shrinkage stress tests with 12mm³ and 24mm³, the comparison between the different composites, as well as the values for Young's modulus are described in Table 2.

Table 2- Shrinkage stress (in MPa) for 12 and 24mm³ increments, as well as Young's modulus (GPa) - Mean (standard deviation)

Time	20s		300s		
Group/ volume	12mm ³	24mm ³	12mm ³	24mm ³	Young's modulus
ADM	0.21 (0.02)ADa	0.23 (0.02)Aa	0.43 (0.03)BCa	0.51 (0.03)ABb	10.3 (1.4)BE
FBP	0.23 (0.03)ABa	0.29 (0.02)ABa	0.43 (0.03)BCa	0.49 (0.4)ABa	17.2 (1.1)D
FBF	0.28 (0.02)BCa	0.43 (0.03)Db	0.53 (0.04)Da	0.72 (0.05)Db	8.0 (0.3)A
SDR	0.20 (0.01)Aa	0.25 (0.02)ABa	0.39 (0.02)ABa	0.45 (0.03)Aa	8.6 (0.4)AB
TBF	0.17 (0.02)Aa	0.32 (0.03)BCb	0.33 (0.03)Aa	0.55 (0.02)BCb	12.4 (1.0)C
XB	0.31 (0.01)Ca	0.51 (0.03)Eb	0.52 (0.02)Da	0.77 (0.05)DEb	10.8 (0.7)CE
XF	0.21 (0.02)ABa	0.36 (0.04)Cb	0.38 (0.03)ABa	0.60 (0.04)Cb	21.6 (1.4)F
Z3F*	0.53(0.03)Ea	0.79 (0.07)Fb	0.88 (0.04)Ea	1.12 (0.03)Fb	8.3 (1.0)A
Z3XT*	0.27 (0.02)BCDa	0.52 (0.02)Eb	0.47 (0.02)CDa	0.83 (0.04)Eb	17.8 (1.7)D

*- Conventional (non-bulk fill) composites

Capital letters mean statistically significant differences between rows in the same column (inter groups), $p \leq 0.05$.

Lower letters mean statistically significant difference between columns within the same row (intra group), regarding the different evaluated times (20s – 12mm³ versus 24mm³; and 300s - 12mm³ versus 24mm³), $p \leq 0.05$.

Considering 12mm³ (Table 2), after 20s, all bulk fill composites except FBF and XB were similar. Only SDR and TBF generated lower stress comparing with the conventional

Z3XT. After 300s, SDR, TBF and FBP generated the lowest stress, followed by the other bulk fill composites, which were similar to Z3XT. Z3F generated the highest stress values for all times.

Considering 24mm^3 (Table 2), Z3F generated the highest stress. After 20s, all bulk fill composites, except XB, generated lower stress values than Z3XT. After 300s, SDR, FBF and ADM generated the lowest stress, followed by TBF and XF. After the volume increase, only ADM, FBP, and SDR generated similar values (20s), independently of the material volume (Table 2). After 300s, FBP and SDR presented similar values regarding the different increment volumes.

Figure 3 shows the development of the shrinkage stress for the different composites. All bulk fill composites showed slower stress development when compared with their regular counterparts (Z3XT or Z3F). ADM showed the slowest stress development.

Figure 3 – Stress development (in MPa) among the different composites (24mm^3).

Considering the Young's modulus, flowable composites (SDR, FBF, XB, and Z3F) presented the lowest values, followed by some high viscosity bulk fill composites (ADM, TBF). The high viscosity composites Z3XT, FBP, and XF presented the highest elastic modulus among the evaluated composites.

Figures 4 and 5 shows the linear regression between Young's modulus and filler content for high and low viscosity composites respectively. It can be observed the lack of correlation for high viscosity composites ($R^2=0.0636$) and a strong correlation for low viscosity composites ($R^2=0.9756$). In addition, a linear regression analysis was performed considering Young's modulus and shrinkage stress (Figures 6 and 7), and no correlation was observed for any of the composite groups.

Figure 4- Young's modulus (GPa) x filler content (%) for high viscosity composites.

Figure 5- Young's modulus (GPa) x filler content (%) for low viscosity composites.

Figure 6- Shrinkage Stress (MPa) x Young's modulus (GPa) for high viscosity composites.

Figure 7- Shrinkage Stress (MPa) x Young's modulus (GPa) for low viscosity composites.

4. DISCUSSION

Polymerization stress generated due to composites inherent shrinkage during light curing has been subject of researches for a long time [21] since stresses that exceed adhesive resistance can lead to gap formation in the adhesive interface [22]. Therefore, the ideal composite should generate the lowest shrinkage stress as possible, capable of ensure a better seal [23].

In order to allow larger increments insertion, the molecular basis of bulk fill composites was modified by incorporation of stress relievers and monomers with higher molecular weight (low molecular weight monomers promote higher number of double bonds per unit of weight, allowing higher degree of conversion, but also leads to higher shrinkage and shrinkage stress)[10,13-16]. One may question about the organic and inorganic matrixes of these composites since “conventional” composites sometimes shares the same composition but, unfortunately, the proportion between the different monomers and fillers is not reported by the manufacturers [10,13,15,21].

In the same way, differences in filler content (such as when comparing high and low viscosity composites) might play an important role at the volumetric shrinkage (higher stress

due to a higher amount of organic content/lower filler content is expected in low viscosity composites). Nevertheless, a lower Young's modulus may allow stress dissipation during polymerization process, thus reducing the stress with bigger increments [18,19].

Considering high viscosity composites with 12mm³ of material after 300s, TBP and FBP generated lower stress, followed by the other bulk fill composites, which presented values similar to the control group Z3XT (Table 2). For low viscosity/flowable composites, SDR generated the lowest stress values, followed by FBF and XB. The low viscosity control group (Z3F) generated the highest shrinkage stress.

In general, high viscosity bulk fill composites generated lower shrinkage stress than low viscosity bulk fill composites as stated by other authors [24]. The only exception was SDR, which generated similar stress when compared with high viscosity bulk fill composites although being flowable. Such results can be explained by the presence of a modified UDMA (monomer with high molecular weight - 849 g/mol), claimed to reduce shrinkage and shrinkage stress [25]. It is interesting to note that all bulk fill composites (high and low viscosities) generated similar or lower stresses when compared with the high viscosity control group (Z3XT).

Since the stress is dependent on the composite volume, being higher with bigger increments [22], it is important to test how the volume increase impacts the generated shrinkage stress [26]. Increased volume (24mm³) resulted in increased tension for the evaluated composites. All bulk fill composites with 24 mm³ generated lower or similar (XB) shrinkage stress when compared with Z3XT after 300s (Table 2). SDR, FBP and ADM generated the lowest stress while Z3F generated the highest stress among all tested composites.

In addition, after 300s, SDR, FBP and ADM, with 24mm³, showed values similar to Z3XT with 12mm³; and FBP and SDR generated similar tensions for both 12 and 24mm³ (Table 2). Such results demonstrate a great capability of bulk fill composites in dealing with shrinkage stress generation even in big increments, as previously reported [15,27].

It is important to note how the increase in volume affected the different classes of composites. Z3XT showed the biggest proportional increase in shrinkage stress while for high viscosity bulk fill composites, the lower volumetric shrinkage might have prevented a bigger increase in stress generation. Besides showing stress values comparable with other bulk fill composites, XF and FBF showed a big increase in stress. For XF, the higher Young's modulus (21.6±1.4GPa)(Table 2), reported also in the literature [28], might have had impact over the polymerization stress for XF (24mm³) due to a sooner development of the

composite viscosity during light curing, leading to a faster stress development when compared with some other high viscosity bulk fill composites (Table 2 and Figure 3). For TBF, the incorporation of 24% prepolymerized fillers increases the filler content amount, but it still might not be able to reduce polymerization shrinkage.

Young's modulus can contribute for a better stress distribution when composites' volumetric shrinkage are similar. It can be noted that all flowable composites showed similar Young's modulus, being lower than high viscosity composites. This lower Young's modulus might explain why they showed a slightly better stress distribution after the increase in the increment volume when compared with Z3XT (Table 2). The higher Young's modulus in high viscosity bulk fill composites might be compensated with a lower polymerization shrinkage, helping to lower stress generation as observed in the present study. It is interesting to note how ADM (10.3 ± 1.4 GPa) and TBF (12.4 ± 1.0 GPa) presented lower Young's modulus when compared with other high viscosity composites. For ADM, probably the ORMOCER based organic matrix is responsible for a more flexible polymer, despite the high filler content. For TBF, the incorporation of 24% prepolymerized fillers increases the filler content without decreasing the elasticity as observed by other authors [28,29].

Considering shrinkage stress development, it could be observed a rapid increase during the first 10s of light curing (total of 20s), followed by a slower increase until the LED light being turned off. The fast subsequent cooling of the composite might be responsible for a second shrinkage peak, as reported by other authors [15,25,30]. Shrinkage stress development seems to be slower in bulk fill composites when compared with conventional resins. This can be observed in Figure 3, in which stress development in bulk fill composites took longer to develop when compared with their regular counterparts. This is especially true when comparing bulk fill composites within the same viscosity (i.e. high viscosity bulk fill composites with lower elastic modulus: ADM and TBF, showed slower stress development). This is important because a slower stress generation allows a better tension distribution and may contribute for the bonding integrity since the material has more time to accommodate the shrinkage stress before the elastic modulus (composite stiffness) start to increase [31,32].

In addition, after 200s, the stress curve is flat for all composites, showing that the major part of the shrinkage develops during the first minutes. This fact can be explained by the increase in the composite rigidity and decrease molecular mobility overtime, even in the presence of free radicals, avoiding further polymerization[32] and explaining the current

option for assessing the shrinkage stress up to 5 minutes instead of several hours as observed by other authors [4,24,30].

In the present study, authors performed correlation tests between elastic modulus and filler content. No correlation was observed for high viscosity composites and strong correlation was observed for low viscosity resins (Figures 4 and 5) besides strong correlation being reported by other authors [30,31]. The low correlation between high viscosity composites might have happened due to ADM and TBF relatively lower elastic modulus when compared with their filler content as discussed previously.

In addition, no correlation was observed between shrinkage stress and Young's modulus for any of the composite's groups (Figures 6 and 7). Such results are in accordance with other authors [25]. This might be explained by Z3XT high volumetric shrinkage and by the use of monomers with higher molecular weight (AUDMA, UDMA and 1, 12-dodecane-DMA) in FBP (Figures 4 and 5). Such results demonstrate how the volumetric shrinkage in fact have a fundamental role on the shrinkage stress generation [25,31]. This statement supports the present study's results since all flowable composites (with lower filler content) are expected to present higher shrinkage and generate higher shrinkage stress [24]. SDR group consist in an outlier as discussed before and as previously reported [25].

Although having benefits that might reflect in easier and faster cavity restorations, bulk fill composites still requires further studies in order to assess the influence of their properties on the long-term maintenance of internal and marginal adaptation. It will be also important to assess the interaction between bulk fill composites and tooth structure regarding adaptation, cusp deflection, among other factors

The present results show, in general, a better behavior for bulk fill composites regarding the shrinkage stress generation, mainly when bigger increments are used. Nevertheless, it is important to note that despite being classified as bulk fill composite resins, the different tested materials can show very different behavior, not only regarding the different classifications (low and high viscosity) as would be expected. This said, further tests are advised in order to clarify to the clinician the best indication for each composite.

5. CONCLUSION

Considering the limitations of the present study, it is possible to conclude that bulk fill composites present very heterogeneous behavior, which is related to their composition (monomers and filler content).

In addition, it can be concluded that:

- Bulk fill resin composites present equal to lower shrinkage stress generation when compared with conventional composites, mainly with bigger increments.
- Bulk fill composites show a wide range of elastic modulus values, but usually similar to “regular” composites
- Volumetric shrinkage seems to be more important than Young’s Modulus for polymerization stress development.

CONFLICTS OF INTEREST:

none

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2.2. ARTICLE 2

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Polymerization shrinkage, microhardness and depth of cure of bulk fill resin composites

Bulk fill composites polymerization properties

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CLINICAL RELEVANCE STATEMENT

Bulk fill composites can be used in large increments (adequate depth of cure up to at least 4.5mm), with similar to lower volumetric shrinkage when compared with conventional composites.

ABSTRACT

The present *in vitro* study assessed the polymerization shrinkage, Knoop microhardness and depth of cure of 9 different composites. The tested composites can be divided into 2 groups: low viscosity: Filtek Bulk Fill Flowable (FBF), Surefill SDR flow (SDR), Xtra Base (XB) and Filtek Z350XT Flowable (Z3F - control); and high viscosity: Filtek Bulk Fill Posterior (FBP), Xtra Fill (SF), Tetric Evo Ceram Bulk Fill (TBF), Admira Fusion x-tra (ADM) and Filtek Z350XT (Z3XT- control). Polymerization shrinkage was assessed with a X-Ray micro computed tomography / μ -CT machine, scanning 64mm³ samples (n=8 for each composite) before and after 20s curing with a 1550mW/cm² LED light curing unit; and quantifying the volumetric differences after image processing and analysis using CTanalyzer. Knoop microhardness and Depth of cure was performed with a microhardness tester, with 0.5mm/min during 30s. For each group, 8 samples had their longitudinal microhardness assessed right after 20s light curing, with 3 readings per depth (separated by 400 μ m) at every 0.5mm. All flowable composites showed lower values for surface microhardness when compared with high viscosity resins. Z3XT showed the highest microhardness among the tested composites, followed by XF, FBP/TBF and ADM. For depth of cure (80% of initial microhardness), Z3XT and Z3F showed lower depth of cure when compared with the bulk fill composites. All bulk fill composites presented depth of cure higher than 4.5mm, being even higher for low viscosity ones. All high viscosity bulk-fill composites showed less shrinkage when compared with the conventional composites (Z3XT and Z3F). All flowable bulk-fill composites showed shrinkage results similar to conventional composites. Bulk fill composites showed similar or lower volumetric shrinkage when compared with “regular” composites, while ensuring proper depth of cure up to at least 4.5mm. Nevertheless, the surface microhardness is widely variable between the composites and they should be assessed carefully in a case-by-case basis, especially for posterior restorations.

Keywords: Composite materials, Biomechanics, Restorative materials, Biomaterial(s), Operative dentistry, Micro-computed tomography

1. INTRODUCTION

Resin composite adhesive restorations have become more popular with the development of new adhesive and composite systems, as well as new filling techniques.⁽¹⁻³⁾ The polymerization shrinkage, an inherent occurrence to resin based materials, can generate fails at the adhesive interface. The amount of shrinkage is dependent on the material composition and volume,⁽⁴⁾ and the pursuit for low shrinkage composites exists for a long time but, since the polymerization process is complex, laboratorial and clinical tests with new composite formulations and insertion techniques are necessary. Besides the increase in composites filler content, another attempt to reduce the polymerization shrinkage was the substitution of Bis-GMA for other monomers like silorane. This modification resulted in a low shrinkage composite but with inadequate mechanical properties.⁽⁵⁾

Recently, with the development of bulk fill composites, the preconized and time costing incremental technique could be substituted for a bulk increment technique. For this, manufacturers claim that the composite is able to control the polymerization process as well as ensure a proper depth of cure even when bigger resin composite increments are used. In order to allow larger increments insertion, the molecular base of these composites was changed by reduction or substitution of Bis-GMA, resulting in a lower viscosity monomer, and/or using monomers with higher molecular weight, usually based on Bis-EMA, TEGDMA, EBPDMA and UDMA monomers.⁽⁶⁾ In addition, incorporation of stress relievers and changes in filler content also helps to control the polymerization shrinkage.⁽⁶⁻¹⁰⁾ Clinically, the volumetric shrinkage can be related with cusp deflection, especially in cavities with high compliance (i.e. low thickness walls) in which the composite shrinkage can be directly reflected on the cavity dimensions,⁽¹¹⁾ as well as in marginal sealing maintenance.^(12,13)

Volumetric shrinkage is a tridimensional phenomenon although being usually assessed through bidimensional methods (and conversion/estimation of tridimensional changes).^(14,15) With the development of new technologies such as x-ray micro computed tomography/u-CT, tridimensional analysis of the volumetric shrinkage might be simpler and reliable.⁽¹⁶⁾

Bulk fill composites can be subdivided into two groups: the materials that can be exposed to the oral environment (usually high viscosity), with greater mechanical properties; and those that should be used as a base/liner (usually low viscosity/flowable), in which the manufacturer recommends a capping layer with conventional resin composite. These characteristics define its indications and different techniques in clinical practice, and can be

partially addressed by mechanical tests. In a clinical situation, despite polymerization shrinkage, adequate depth of cure also consists in a major concern for use of big increments. In addition, use of bigger increments usually are associated with a higher C-factor, which can increase shrinkage stress development.⁽¹⁷⁾ Manufacturers' used different approaches to ensure a proper polymerization in deep cavities such as incorporation of new and more reactive photoinitiators, reduction of composite's translucency (through filler content changes such as increase in particles size or reduction of the mismatch between fillers and organic matrix refractive indexes).^(18,19) The increased translucency for some composites can be observed during clinical use and ensuring adequate physico-mechanical properties⁽²⁰⁾ and long term maintenance of restoration.

The objectives of the present study were to assess the polymerization shrinkage using a tridimensional method (μ -CT), microhardness and depth of cure of different composites. The null hypotheses tested were: 1) there would be no differences in surface microhardness when comparing the different composites; 2) Bulk fill composites would not present higher depth of cure than regular composites and 3) Bulk fill composites would not present lower polymerization shrinkage than regular composites;

2. METHODS AND MATERIALS

2.1. STUDY DESIGN AND MATERIALS

Resin composites were evaluated in 9 levels (nine different materials)(Table 1), having as response variables: microhardness and depth of cure through microhardness test, and volumetric polymerization shrinkage through μ -CT analysis.

Table 1 – Different groups with respective composition and manufacturers.

Group	Restorative material	Composition (organic matrix, filler weight)	Viscosity, type and increment size	Manufacturer
ADM	Admira Fusion x-tra	Ormocer resin, 84% filler	Regular, bulk fill, up to 4mm	VOCO, Cuxhaven, Germany
FBP	Filtek Bulk Fill Posterior	AUDMA, UDMA and 1, 12-dodecane-DMA, 76.5% filler	Regular, bulk fill, up to 5mm	3M ESPE, St Paul, MN, USA
TBF	Tetric Evo Ceram Bulk Fill	Bis-GMA, UDMA, 78% filler	Regular, bulk fill, up to 4mm	Ivoclar Vivadent, Schaan, Liechtenstein
XF	X-tra Fil	Bis-GMA, UDMA, TEGDMA, 86% filler	Regular, bulk fill, up to 4mm	VOCO, Cuxhaven, Germany
Z3XT	Filtek Z350XT	Bis-GMA, Bis-EMA, UDMA, TEGDMA, 82% filler	Regular, conventional, up to 2mm	3M ESPE, St Paul, MN, USA
FBF	Filtek Bulk Fill Flowable	UDMA, BISGMA, Bis-EMA, Procrylat resin, 64.5% filler	Flowable, bulk fill, up to 4mm	3M ESPE, St Paul, MN, USA
SDR	Surefil SDR flow	Modified UDMA, EBPADMA, TEGDMA, 68% filler	Flowable, bulk fill, up to 4mm	Dentsply Sirona, York, PA, USA
XB	X-tra Base	UDMA, Bis-EMA, 75% filler	Flowable, bulk fill, up to 4mm	VOCO, Cuxhaven, Germany
Z3F	Filtek Z350 flow	Bis-GMA, Bis-EMA, TEGDMA, 65% filler	Flowable conventional, up to 2mm	3M ESPE, St Paul, MN, USA

2.2. METHODS

2.2.1. Microhardness and Depth of Cure

For the Knoop microhardness test, eight specimens for each group were achieved through insertion of composite into a metallic matrix designed in a CNC lathe (Sherline 2000, Sherline, Vista, CA, USA) controlled through the Match 3 software (Newflanged solutions, Livermore Falls, ME, USA), with 10mm length and a central trapezoidal-shaped groove (Figure 1A).

Figure 1- Metallic matrix for longitudinal microhardness test. A- Details of the bi-part matrix; B- Resin composite sample and metallic matrix in position.

After composite insertion into the groove, a polyester strip was positioned over the top surface to standardize the surface roughness and a metallic cover was put in position (Figure 1B). The composite excess was removed and the specimen was light cured during 20s through the small window with a 1550mW/cm² irradiance (31J/cm² total irradiation) LED light curing unit (LED Blue Star 3, Microdont, São Paulo, SP, Brazil), assessed with a radiometer. A PVS matrix was used to standardize the light curing unit in a perpendicular position and in contact with the polyester strip.

After light curing, the metallic cover and polyester strip were removed. The non-polymerized composite was removed with aid of a #12 scalpel blade and the top surface of the specimen was analyzed with a Knoop microhardness tester (Micromet 6040, Buehler, Lake Bluff, IL, USA) in all its longitudinal extension through 3 surface readings for each 0.5mm, separated by 400 μ m. The final result for each depth was calculated as the 3 readings mean value. The first reading (surface) was considered as 0.5mm from the

with 648X512 resolution. The slice thickness was determined as 16 μ m for a scan time of 20 minutes.

Four samples were assessed at each scan. Specimens were prepared inside a x-ray dark room and were transported to the μ -CT chamber in a dark storage in order to avoid light interference.

The first scanning had as goal to assess the total material volume before polymerization. After the first scanning, the specimens were light cured during 20s with the LED light curing device, and the polymerized samples were scanned again, as described before. All obtained images were rendered in specific softwares: CT-Analyzer (CTAn) and CT-volume (CTVol)(Bruker, Kontich, Belgium); to obtain tridimensional specimen's reproduction (Figure 2B). The volumetric shrinkage was determined as the percentage difference between the initial (before light curing) and final volume (after light curing).

2.3. STATISTICAL ANALYSIS

After Shapiro-Wilk normality test, all data were analyzed through one-way Anova followed by Tukey's test. For all the statistical analysis, 5% was adopted as significance level ($p \leq 0.05$). In addition, linear correlation tests were performed to associate depth of cure, filler content and/or volumetric shrinkage for both high and low viscosity composites.

3. RESULTS

All flowable composites showed lower values for surface microhardness (FBF presented the lowest one). For high viscosity composites, Z3XT presented the highest microhardness values, followed by XF, FBP/TBF, and ADM. For depth of cure (80% of initial microhardness), conventional composites showed lower depth of cure when compared with the bulk fill composites. All bulk fill composites presented depth of cure values higher than 4.5mm. SDR and XB showed the highest values (Table 2).

Table 2- Surface microhardness (Knoop) and depth of cure (80% of initial microhardness).

Group	Surface microhardness (g/mm ²)	Depth of cure (80%)(mm)
ADM	37.4 (5.15)D	5.4 (0.6)BC
FBP	50.0 (2.4)C	5 (0.5)C
TBF	50.9 (5.2)C	4.9 (0.4)C
XF	74.3 (10.7)B	5.4 (0.7)BC
Z3XT	89.4 (6.8)A	2.6 (0.2)E
FBF	16.2 (2.3)E	5.6 (0.3)BC
SDR	22.0 (2.1)E	6.9 (0.4)A
XB	31.9 (2.7)D	6.1 (0.7)B
Z3F	33.3 (3.1)D	3.6 (0.2)D

Different letters means statistically significance difference between rows in the same column (inter-groups comparison)($p \leq 0.05$)

The initial measured volume and shrinkage after light curing are listed in table 3. All groups presented similar initial volumes, with low standard deviation. All high viscosity bulk fill composites showed lower volumetric shrinkage when compared with Z3XT and Z3F. All flowable bulk fill composites showed results similar to Z3XT and Z3F. Z3F presented the highest shrinkage values while XF presented the lowest shrinkage values.

Table 3 – Initial volume (mm³) and volumetric shrinkage (%).

Group	Initial volume (mm ³)	Volumetric Shrinkage (%)
ADM	61.2 (2.0)A	1.2 (0.2)AC
FBP	61.5 (1.1)A	2.2 (0.5)B
TBF	61.1 (1.7)A	1.7 (0.1)BC
XF	62.2 (1.1)A	0.8 (0.4)A
Z3XT	60.9 (1.1)A	3.1 (0.6)D
FBF	60.2 (2.2)A	3.3 (0.6)DE
SDR	61.9 (1.5)A	3.4 (0.6)DE
XB	63.3 (2.2)A	3.1 (0.2)DE
Z3F	62.5 (1.2)A	3.8 (0.2)E

Different letters means statistically significance difference between rows (inter-groups comparison)($p \leq 0.05$)

In the Figures 3-6 it is possible to observe the correlation between different factors. Figure 3 shows a medium correlation between all groups considering microhardness and filler content ($R^2=0.5708$).

Figure 3 – Linear correlation between Microhardness (g/mm²) and Filler Content (%).

In the Figure 4, it is possible to observe a lack of correlation between depth of cure and filler content for high viscosity composites when all groups are considered ($R^2=0.0085$). Nevertheless, a strong correlation is observed when only high viscosity bulk fill composites are considered ($R^2=0.8397$).

Figure 4 – Linear correlation between Depth of Cure (mm) and Filler Content (%) for high viscosity composites.

Figure 5 shows the lack of linear correlation between depth of cure and filler content for low viscosity composites ($R^2=0.2169$).

Figure 5 – Linear correlation between Depth of Cure (mm) and Filler Content (%) for low viscosity composites.

Figure 6 shows a strong correlation between filler content and volumetric shrinkage for all groups ($R^2=0.6918$), which is even stronger when Z3XT is excluded ($R^2=0.9081$).

Figure 6 – Linear correlation between Filler Content (%) and Volumetric Shrinkage (%).

4. DISCUSSION

Changes in both organic and inorganic matrix can have influence on composites mechanical and physical properties. Despite being considered as bulk fill composites, these materials show very heterogeneous behavior, being important to assess their properties.⁽²³⁾

Very heterogeneous results could be observed for the surface microhardness test, even when comparing the high viscosity bulk fill composites, indicated to be exposed at the occlusal surface. Conventional Z3XT (89.4 ± 6.8) and XF (74.3 ± 10.7), showed the highest microhardness values and among the highest filler contents according to the respective manufacturers' information. TBF (50.9 ± 5.2) and FBP (49.6 ± 2.4) showed similar microhardness values and present similar filler contents (78% and 76.5% respectively). Interestingly, despite also having a high filler content (84%), ADM presented lower microhardness values (37.4 ± 5.1). Such differences might be explained by the different fillers size and distribution, as well as due to the differences in the Ormocer organic matrix. The results for Z3XT and TBF are similar to the ones reported by Rodriguez et al.⁽²⁴⁾

As expected, microhardness' results for lower viscosity composites were lower when compared with high viscosity composites,^(19,25) with Z3F (33.3 ± 3.1) and XB (31.9 ± 2.7) showing the highest values, followed by flowable SDR (22.0 ± 2.1) and FBF (16.2 ± 2.3). Despite presenting similar filling content as FBF and SDR, Z3F (65%) showed similar microhardness values as XB, which presents a higher filler content (75%). This might be explained by the use of nanoparticles and nanoclusters in Z3F. The presented results clearly show the manufacturers' indications for clinical use. The high viscosity composites showed very heterogeneous results, but definitely higher than the flowable composites, which might need a capping layer. The heterogeneous values observed for high viscosity composites can rely partially on the lower elastic modulus observed for some of the bulk fill composites, associated with the different filler contents and organic matrix composition.⁽²⁵⁻²⁸⁾ This said, it might be important to consider each composite according to each clinical indication.

An increase in microhardness values is expected as the filler content increases.^(19,24,25,27,29) In the present study, it was possible to observe a medium correlation between microhardness and filler content ($R^2=0.5708$)(Figure 3). As observed in figure 3, the correlation probably was not higher due to the different organic matrix of ADM, and to the high filler content of XB besides being a low viscosity composite, presenting a high microhardness value for this class of composites (Table 2). Since the different composites showed very different values for microhardness, the first null hypothesis was rejected.

Using the microhardness test, it was also possible to determine the composites' depth of cure, through longitudinal test, being considered adequate when values equal or higher than 80% are achieved (comparing with the first readings mean values/ upper surface readings).^(22,30) All the tested bulk fill composites showed adequate polymerization at least up to 4.5mm; being even higher for low viscosity bulk fill composites (at least 5mm). Conventional composites presented adequate depth of cure up to 2.6mm (Z3XT) and 3.6mm (Z3F), which explains why they should not be used in big increments. Similar results were described in the literature.^(19,22,24,28-31) Since all bulk fill composites showed adequate polymerization at least up to 4.5mm (versus 3.5mm from conventional Z3F and 2.5mm from Z3XT), the second null hypothesis was rejected (difference between conventional and bulk fill composites).

Another interesting finding with the longitudinal microhardness is the increase in the microhardness values up to 2mm before starting to decrease. This was reported before in the literature and, since the oxygen inhibited layer corresponds to 20-50µm and was avoided in the present study (polymerization in contact with polyester strips), such behavior can be related with the shrinkage of the composite towards the center in non-bonded models.^(6,19)

Except for the TBF, which relies on a new dibenzoyl germanium compound (Ivocerin), which absorbs light between 370 and 460nm and is claimed to be more reactive, the other bulk fill composites shows the same camphorquinone/amine initiator system, probably relying in changes in the filler contents and higher translucency for improvements on the depth of cure.^(19,23,26,27) A more translucent composite can be achieved through reduction in the filler content, use of bigger particles size and study of the interaction between the fillers and organic matrix refractive indexes.^(18,19) The increased translucency of the bulk fill resin composites can be observed in clinical situation, in which the necessity of a capping layer comes from both mechanical and esthetical properties.

Since manufacturers could improve the depth of cure through changing the inorganic matrix,^(24,32) a correlation test between the filler content and the depth of cure was performed. Considering high viscosity composites, it is possible to observe a strong correlation when Z3XT is excluded ($R^2=0.8397$)(Figure 4). As expected, Z3XT is an outlier in this comparison since it is not supposed to be light cured in big increments due to a limited depth of cure (Table 2). Performing the same analysis for low viscosity composites, no correlation was observed ($R^2=0.2169$)(Figure 5).

Another important question when polymerizing big increments consist in the polymerization shrinkage. Usually, volumetric shrinkage is assessed through bidimensional

measurements and conversion of values estimating the volume.^(14,15) With the use of new technologies such as μ -CT, it is possible to assess the real volumetric shrinkage of the composites.⁽¹⁶⁾ It can be observed that the conventional composites and the flowable bulk fill composites showed similar volumetric shrinkage (around 3.2%). The highest value was observed for the low viscosity Z3F ($3.8 \pm 0.2\%$), while the high viscosity bulk fill composites showed lower but heterogeneous results, ranging from $0.8 \pm 0.4\%$ (XF) up to $2.2 \pm 0.5\%$ (FBP). Such results are in agreement with the literature, which reported values between 1-3% for high viscosity composites and up to 6% for low viscosity ones.⁽³⁰⁾ In a recent study by Yu et al.,⁽³¹⁾ the results for SDR and TBF were similar to the results of the present study despite the different methods, validating the use of μ -CT. It is important to note that even some bulk fill composites show similar shrinkage values when compared with the conventional Z3XT ($3.1 \pm 0.6\%$), this is not necessarily related with an increased shrinkage stress since it depends also on the elastic modulus and development, polymerization kinetics, among other factors.^(30,33-35)

It is interesting to observe that the flowable bulk fill composites showed similar values when compared with the conventional high viscosity composite. This fact demonstrates that the mechanisms used in the bulk composites in order to reduce the shrinkage, such as introduction of monomers with higher molecular weight (reducing the number of reactive sites per volume)⁽³⁶⁾ and increase in filler content, were able to effectively reduce the shrinkage.⁽³⁷⁾ Using the same techniques and further increasing the filler content resulted in even lower volumetric shrinkage as observed for the high viscosity bulk fill composites, especially for ADM, TBF and XF. In fact, the results of the present study show a strong correlation between filler content and volumetric shrinkage ($R^2=0.6918$)(Figure 6). Similar results were reported by Al sunbul et al. (2016).⁽³⁷⁾ Z3XT can be considered as an outlier and this can be explained by the lower shrinkage observed for all high viscosity bulk fill composites regardless the filler content (Table 3) and, repeating the linear regression analysis without Z3XT resulted in $R^2=0.9081$. Since the shrinkage values varied widely, the third null hypothesis was also rejected.

The use of μ -CT for tridimensional volumetric shrinkage assessment consist in a reliable and simpler test when compared with other methods such linear shrinkage assessment and estimation of the tridimensional changes, dilatometer based methods, among others. In addition, the use of a Teflon matrix instead a tooth cavity avoids possible interferences ("bonding") between the composite and the adjacent walls/structures.

All bulk fill composites presented equal to lower polymerization shrinkage when compared with conventional composites. These properties, associated with the increased depth of cure are interesting for clinical application, with possibility of easier and faster restoration placement. Nevertheless, the low microhardness values for some composites can be concerning in some clinical situations, especially regarding occlusal cavities. Thus, further studies are necessary assessing the restorative properties of such composites.

5. CONCLUSION

Considering the limitations of the present study, it is possible to conclude:

- The surface microhardness is widely variable between the tested composites. No bulk fill composite achieved the same surface microhardness as Filtek Z3XT.
- All tested Bulk fill composites showed proper depth of cure up to at least 4.5mm being indicated for bulk placement, and presenting higher depth of cure than conventional composites.
- All tested bulk fill composites showed similar or lower volumetric shrinkage when compared with conventional composites.

CONFLICTS OF INTEREST

The authors report no conflicts of interest.

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3. DISCUSSION

3. DISCUSSION

The results of the present study show the heterogeneity of the bulk fill composites, besides being considered as only one group of material (ENGELHARDT et al., 2016). Bulk fill composites can be divided into two groups: high viscosity - composites that can be exposed to the oral environment, with greater resistance to physico-mechanical challenges (such as masticatory forces, abrasion, acid challenges, among others); and low viscosity – composites that should be used as a base material in association with a cover layer with conventional resin composite (ILIE et al., 2013). These characteristics define its indications and different techniques in clinical practice.

All bulk fill composites showed an increased depth of cure (over 4mm for all materials), being very interesting from a clinical aspect since it ensures a shorter clinical time and a less sensitive restorative technique while keeping adequate polymerization reliability with equal or lower polymerization shrinkage/shrinkage stress when compared with regular composites. Another common finding was the similar to lower volumetric shrinkage and shrinkage tension when compared with regular composites (Z3XT and Z3F). Despite that, the elastic modulus and microhardness were widely variable, and that might result in different clinical behavior, being important to carefully address the long-term performance of those restorations in a clinical situation.

Microhardness tests are important to predict the mechanical behavior of the composite surface against the oral cavity erosive and abrasive challenges (CHANG et al., 2013; RODRIGUEZ et al., 2017). Usually, high viscosity composites should present higher values in order to resist to mechanical occlusal forces without losing its shape and/or volume. As expected, there was a clear difference between low and high viscosity composites, with the former being more adequate to be used as a base material. Nevertheless, when assessing high viscosity composites, it was interesting to observe the wide range of microhardness values, strongly correlated with the filler content. This said, it is important to further assess those composites after artificial aging using mechanical and abrasion resistance tests.

One of the biggest advantages of bulk fill composites consist in the possibility of use bigger increments despite the concerns about polymerization shrinkage, shrinkage stress and adequate polymerization properties. Using the same microhardness test setup, it is also possible to assess composites' depth of cure in an easy and reproducible way (ALRAHLAH et al., 2014; BOUSCHLICHER et al., 2004; EL-DAMANHOURY, PLATT, 2014). Despite the

fact that microhardness tests do not consist in a direct degree of conversion test (such as Fourier Transformed Infra-Red Spectroscopy- FTIR for example), they show strong correlation with this test, being about 2.5 times more sensitive (BOUSCHLICHER et al., 2004).

As observed in the present results, it is safe to use increments up to 4-5mm without compromises of light curing efficiency, showing that the mechanisms adopted in order to increase the light transmittance and reactivity of monomers were successful, as stated by other authors (ALRAHLAH et al., 2014; BOUSCHLICHER et al., 2004; EL-DAMANHOURY, PLATT, 2014; EL-SAFTY et al., 2012; FLURY et al., 2012; ILIE et al., 2013). The increased light transmittance was achieved through either organic content changes (use of different monomers), use of different filler content, size, shapes and distributions; and/or use of new photoinitiators such as Ivocerin (on TBF), claimed to be more reactive than camphorquinone to visible light due to a higher quantum efficiency of light induced cleavage (AZZOPARDI et al., 2009; BUCUTA, ILIE, 2014; DIONYSOPOULOS et al., 2016; ENGELHARDT et al., 2016; ILIE et al., 2013; ILIE, STARK, 2014; LEE, 2008). Important to note, that results can be different after storage and/or using a different matrix. This said, immediate microhardness readings were preferred to facilitate analysis and workflow and, although the results might be lower than after 24 hours storage for example, they follow the same trend (ALSHALI et al., 2015). The use of a metallic matrix was preferred in order to avoid any light transmission and depth of cure overestimation (PRICE et al., 2016).

Considering use of bulk fill composites in bigger increments, it is important to assess not only the quality of the increment polymerization, but also the shrinkage stress and volumetric shrinkage they generate (AREGAWI, FOK, 2017). The shrinkage stress can be considered a result of the volumetric shrinkage and elastic modulus. With aid of a Universal testing machine ("Tensilometer"), it was possible to measure the shrinkage stress of different composites over time, considering different increment volumes (ISHIKIRIAMA et al., 2013; ISHIKIRIAMA et al., 2012). It was possible to observe a tendency of lower stress generation for bulk fill composites, especially high viscosity bulk fill composites when compared with other groups. Such tendency for high viscosity bulk fill composites was more pronounced as the volume was increased. This probably can be explained by a lower volumetric shrinkage (as observed with the μ CT analysis) and/or relatively lower elastic modulus when compared with conventional high viscosity composites (as observed with the Young's modulus test).

Young's modulus also can be assessed with an universal testing machine through a 3-point bending test device. This test shows the composite stiffness, as a more elastic composite could dissipate/reduce stress at the adhesive interface better than stiff composites inside a cavity (JUNG, PARK, 2017; KIM et al., 2016; LEPRINCE et al., 2014). Young's modulus can also be associate with filler content, and is generally lower for low viscosity composites. Despite this common assumption, it is interesting to observe some high viscosity bulk fill composites (with high filler content) showing low Young's modulus, probably as a way to keep the shrinkage stress under control. The relatively lower Young's modulus for high viscosity composites can be explained by the use of pre-polymerized particles and changes in the organic matrix.

The use of μ CT to assess volumetric shrinkage also proved to be an interesting tool, assessing the tridimensional alterations in a direct way. This setup avoids discrepancies since literature has been reporting volumetric shrinkage based in bidimensional tests, which really depends on the adopted test setup (HAN et al., 2016). In addition, the use of a Teflon matrix instead of tooth structure ensures the composite shrinkage happened freely and was not prevented by any bonding force. The interaction between the results achieved with the universal testing machine (shrinkage stress and elastic modulus) and the μ CT (volumetric shrinkage), shows the reliability of the test setup (Table 1).

Such results could provide the total shrinkage amount, but further studies assessing the volumetric shrinkage in cavities with different C-factors and with bonding agents will be interesting to show the shrinkage direction and to provide a better comprehension of how to avoid gaps at the interface between tooth structure and restorations.

High viscosity bulk fill composites showed less volumetric shrinkage when compared with regular composites and low viscosity bulk fill composites (with similar results).

A strong correlation was found between Volumetric shrinkage, Young's modulus and Shrinkage Stress for the tested composites ($R^2=0.7195$ without consider the conventional composites and SDR). As expected, regular composites can be considered outliers when compared with the bulk fill composites since they were expected to behave very differently due to the lack of already discussed mechanisms to control the polymerization process. Comparing the bulk fill composites, SDR also can be considered as an outlier since it is a flowable composite that, from a polymerization shrinkage point-of-view, shows the same behavior as a high viscosity composite (Table 1).

Table 1 – Linear regression between Volumetric shrinkage (%), Shrinkage stress (MPa) and Young's modulus (GPa).

In general, as the volumetric shrinkage increased (low viscosity bulk fill composites), the elastic modulus was lower and the shrinkage stress could be reduced. Similarly, when the volumetric shrinkage was lower (high viscosity bulk fill composites), the elastic modulus was higher and the shrinkage stress was kept at similar values.

Since bulk fill composites are relatively new on the market, there is still only a few clinical trial reports, especially when we consider the number of currently available bulk fill composites. This said, in vitro tests are very important to predict the clinical performance, especially considering the restorative performance after simulated artificial aging.

The use of base materials with low elastic modulus such as glass ionomer cements, flow composites and bonding agents, or even bulk fill composites could be a strategy to reduce the polymerization shrinkage (PECIE et al., 2013). This suggestion was based on the theory that an elastic material could neutralize some of the stresses generated during light-curing, without compromising the intimate relationship between the restorative material and tooth structures; as well as reducing the composite volume (CARA et al., 2007; KARAMAN, OZGUNALTAY, 2014; NAGEM FILHO et al., 2007). This said, it is also important to test other cavity configurations (with different compliances), and association between base and cover materials in order to assess their interactions and performance.

Considering the limitations of the present research protocol, during the polymerization stress analysis, we adopted a “rigid compliance” device since, despite not having a feedback system, metal plates were used. This said, only the composite specimen and the loading cell could suffer deformations. This configuration could generate higher stress values than the observed in tooth structure since the dentin resilience, cusps deformation, as well as a tridimensional component, are present in the clinical scenario and cannot be reproduced with the universal testing machine. For future studies, the analysis of the tooth/restoration interface considering different tooth structure compliance might be important.

Considering the microhardness test for depth of cure assessment, it consist in a simple and reliable tool. We adopted an immediate assessment of the surface microhardness without polishing or water storage. This approach could generate different microhardness values when compared with the literature, but the depth of cure might be accurate since the whole specimen had the same treatment. The use of u-CT to assess the polymerization shrinkage proved to be a reliable and simpler test when compared with other methods such as linear shrinkage assessment and estimation of the tridimensional changes, dilatometer based methods, among others. In addition, the use of a PTFE matrix instead a teeth cavity avoids possible interferences (“bonding”) between the composite and the adjacent walls/structures.

4. FINAL CONSIDERATIONS

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Bulk fill composites can be used in big increments with similar or lower volumetric shrinkage and shrinkage stress than regular composites, while ensuring proper depth of cure of at least 4.5mm

Bulk fill composites present very heterogeneous mechanical behavior (microhardness), even considering low viscosity or high viscosity groups, being recommended an individual assessment of each material according to the clinical situation.

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ANNEXES

ANNEXE 1

Journal of Dentistry <eesserver@eesmail.elsevier.com>
Reply-To: Journal of Dentistry <JoD@elsevier.com>
To: fabio.rizzante@gmail.com, fabioantonio_7@hotmail.com
Cc: chris.lynch@ucc.ie

Mon, Jun 26, 2017 at 5:08 AM

Ref.: Ms. No. JJOD-D-17-00465
Mechanical properties assessment of Bulk Fill resin composites
Journal of Dentistry

Dear Dr. Rizzante,

Thank you for submitting your manuscript to the Journal of Dentistry. Your paper has now been reviewed by our international experts.

Based on our reviewer(s) recommendations and after careful deliberations, I regret to advise you that we are unable to publish your contribution to the journal. I have enclosed some editorial comments, which you might find useful should you submit your paper elsewhere.

Manuscripts submitted for publication are critically evaluated by the Editorial board as to their scientific importance in the area of research covered. Your manuscript was assessed and was not considered to be of sufficient priority for publication in The Journal of Dentistry.

Although your contribution was not successful on this occasion, we hope that you will continue to consider Journal of Dentistry for future submissions.

Thank you for giving us the opportunity to consider your work.

Yours sincerely

Professor. Christopher D. Lynch
Editor in Chief
Journal of Dentistry

ANNEXE 2

editor@jopdent.org <editor@jopdent.org>

Mon, Oct 30, 2017 at 8:29 AM

Reply-To: editor@jopdent.org

To: fabio.rizzante@gmail.com

October 30, 2017

Dear Author,

The referee comments regarding your manuscript "Polymerization shrinkage, microhardness and depth of cure of bulk fill resin composites" have been received. On the basis of the reviews I regret to inform you that I cannot accept this article for publication. The referee comments are attached and are written to help our colleagues improve on content presentation and/or improve their research techniques. I urge you to accept these criticisms in the spirit in which they are offered.

At the current time, Operative Dentistry receives a large number of outstanding papers. This makes it necessary for us to be extremely conservative in our acceptance of new manuscripts. Even relatively minor errors in protocols, English language usage, reporting of the research or minor contribution to our knowledge base can prevent acceptance of a paper.

It is obvious that much time and effort was spent in creating this manuscript. We are honored that you would submit this paper for consideration by Operative Dentistry and look forward to receiving future papers from you for publication consideration.

Sincerely,

Jeffrey Platt
Editor
Operative Dentistry

ANNEXE 3

1/15/2018

"journal of dentistry" - NLM Catalog - NCBI

NLM Catalog ▼ "journal of dentistry" ✕

Summary Sort by Publication Date

Search results

Items: 5

Filters activated: Journals referenced in the NCBI DBs, Journals currently indexed in MEDLINE. [Clear all](#) to show 50 items.

- ☐ [Journal of dentistry for children \(Chicago, Ill.\)](#)
- 1. American Academy of Pediatric Dentistry.
NLM Title Abbreviation: J Dent Child (Chic)
ISSN: 1551-8949 (Print) ; 1935-5068 (Electronic) ; 1551-8949 (Linking)
[Chicago, Ill.] : American Academy of Pediatric Dentistry, [2003]-
Currently indexed for MEDLINE
NLM ID: 101180951 [Serial]
- ☐ [American journal of dentistry](#)
- 2. NLM Title Abbreviation: Am J Dent
ISSN: 0894-8275 (Print) ; 0894-8275 (Linking)
[San Antonio, TX : Mosher & Linder, Inc., c1988-
Currently indexed for MEDLINE
NLM ID: 8806701 [Serial]
- ☐ [Special care in dentistry : official publication of the American Association of Hospital Dentists, the Academy of Dentistry for the Handicapped, and the American Society for Geriatric Dentistry](#)
- 3. American Association of Hospital Dentists; Academy of Dentistry for the Handicapped; American Society for Geriatric Dentistry; American Dental Association.
NLM Title Abbreviation: Spec Care Dentist
ISSN: 0275-1879 (Print) ; 1754-4505 (Electronic) ; 0275-1879 (Linking)
[Chicago : American Dental Association, c1981-
Currently indexed for MEDLINE
NLM ID: 8103755 [Serial]
- ☐ [Journal of dentistry](#)
- 4. NLM Title Abbreviation: J Dent
ISSN: 0300-5712 (Print) ; 1879-176X (Electronic) ; 0300-5712 (Linking)
Kidlington : Elsevier
Currently indexed for MEDLINE
NLM ID: 0354422 [Serial]
- ☐ [Australian dental journal](#)
- 5. Australian Dental Association.
NLM Title Abbreviation: Aust Dent J
ISSN: 0045-0421 (Print) ; 1834-7819 (Electronic) ; 0045-0421 (Linking)
Sydney : Australian Dental Association
Currently indexed for MEDLINE
NLM ID: 0370612 [Serial]

ANNEXE 4

1/15/2018

oper dent - NLM Catalog - NCBI

 [Summary](#) [Sort by Publication Date](#)**Search results****Items: 2**Filters activated: Journals referenced in the NCBI DBs, Journals currently indexed in MEDLINE. [Clear all](#) to show 6 items.☐ [Operative dentistry. Supplement](#)

1. University of Washington School of Dentistry; Academy of Operative Dentistry; American Academy of Gold Foil Operators.

NLM Title Abbreviation: **Oper Dent** Suppl

ISSN: 0163-3473 (Print) ; 0163-3473 (Linking)

Seattle, WA : University of Washington, School of Dentistry

Currently indexed for MEDLINE

NLM ID: 7805058 [Serial]

☐ [Operative dentistry](#)

2. Academy of Operative Dentistry; American Academy of Gold Foil Operators; University of Washington School of Dentistry.

NLM Title Abbreviation: **Oper Dent**

ISSN: 0361-7734 (Print) ; 1559-2863 (Electronic) ; 0361-7734 (Linking)

Seattle, Univ. of Washington, School of Dentistry.

Currently indexed for MEDLINE

NLM ID: 7605679 [Serial]

ANNEXE 5

DECLARATION OF EXCLUSIVE USE OF THE ARTICLE IN DISSERTATION/THESIS

We hereby declare to be aware that the article “Mechanical properties of Bulk Fill resin composites” will be included in the thesis of the student Fabio Antonio Piola Rizzante and cannot be used by other works/Graduate programs at the Bauru School of Dentistry, University of São Paulo.

Bauru, January 21st, 2018

Fabio Antonio Piola Rizzante

Rafael Francisco Lia Mondelli

Adilson Yoshio Furuse

Ana Flávia Sanches Borges

Gustavo Mendonça

Sérgio Kiyoshi Ishikiriama

ANNEXE 6

DECLARATION OF EXCLUSIVE USE OF THE ARTICLE IN DISSERTATION/THESIS

We hereby declare to be aware that the article "Polymerization shrinkage, microhardness and depth of cure of bulk fill resin composites" will be included in the thesis of the student Fabio Antonio Piola Rizzante and cannot be used by other works/Graduate programs at the Bauru School of Dentistry, University of São Paulo.

Bauru, January 21st, 2018

Fabio Antonio Piola Rizzante

Jussaro Alves Duque

Marco Antônio Húngaro Duarte

Rafael Francisco Lia Mondelli

Gustavo Mendonça

Sérgio Kiyoshi Ishikiriama