



Physical-Mechanical Properties of Bulk Fill Composites Submitted to Biodegradation by *Streptococcus mutans*

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The aim of this study was to evaluate the *Streptococcus mutans* biofilm influence on the roughness (Ra), gloss (GU), surface hardness (KHN) and flexural strength (FS) of high viscosity bulk fill composites. Filtek Bulk Fill (FBF), Tetric N Ceram Bulk Fill (TNC), X-tra fil Bulk Fill (XF) and Filtek Z350 (FZ) were used. Ten discs of each composite were prepared for Ra, KHN and GU and 20 bars for the FS. After 24 h, specimens were polished and initial analyzes performed. Samples were sterilized and subjected to biodegradation for 7 days and final analyzes performed. Representative samples of each group were evaluated in Scanning Electron Microscope. Data were submitted to ANOVA two factors and Tukey test. XF presented the highest values ($p < 0.05$) of Ra before and after biodegradation (0.1251; 0.3100), and FZ (0.1443) the lowest after biodegradation ($p < 0.05$). The highest GU values ($p < 0.05$) were observed for FZ (71.7; 62) and FBF (69.0; 64.6), and the lowest ($p < 0.05$) for TNC (61.4; 53.3) and XF (58.5; 53.5), both before and after biodegradation. For KHN the highest values were obtained by XF (151.7; 106), and the ($p < 0.05$) lowest values for TNC (62.2; 51.8), both before and after biodegradation. The highest values ($p < 0.05$) of FS were observed for FZ (127.6) and the lowest ($p < 0.05$) for TNC (86.9); after biodegradation, XF (117.7) presented the highest ($p < 0.05$) values compared to TNC and FZ." In conclusion, biodegradation increased Ra and decreased GU and KHN for all. Concerning FS, degradation provided a significant decreased value only for FZ.

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Introduction

Due to greater demand for aesthetics in dentistry, the composite resin has been the material of choice for direct restorative treatment. In order to simplify the restorative procedure, reduced clinical time, and decrease polymerization shrinkage stresses and cusp deflection, Bulk Fill composites were introduced into dental market with the appeal of simplifying the restorative procedure by single increment insertion of up to 4 mm depth with an adequate polymerization (1).

According to the manufacturers, these composites require a short activation time due to the modification of the initiation system that includes higher concentrations of conventional photoinitiators and reduced light scattering at the filler-matrix interface by decreasing the filler amount or increasing the filler size (2), providing satisfactory material polymerization.

Despite the evolution of composite resins related to filler content, resin matrix and initiator systems (3), biofilm is the highest challenge for restorative materials on oral cavity. In addition, some enzymes from the class of esterases such as cholesterol esterase and pseudocholinesterase can degrade BisGMA/TEGMA based polymer composite in polymer matrix (4). Moreover, the cariogenic bacteria

from dental biofilm denominated *Streptococcus mutans*, is capable of degrading the resin matrix of composites not just because of their esterase activity but also by producing acids and decreasing the pH environment (5). Further the destructive effect on the polymers, it may affect the materials properties in short or long term (6).

One drawback of the acids produced by the bacteria of the cariogenic biofilm is the softening of the resin matrix leading to increasing surface roughness and reducing hardness and gloss (7). Moreover, the flexural strength is considered an indicative factor about the performance and longevity of the composites when subjected to biological degradation and masticatory forces, influencing the material performance (8).

Although there are several studies in the literature in relation to Bulk Fill composites related mainly to mechanical performance (1-3,9,10), they are not focused on the influence of cariogenic biofilm on the properties of materials. Considering the routine use of composites resins to restore posterior teeth and the potential deleterious effect of biofilm on the surface of restorative materials as shown by the literature (6), it would be essential to evaluate the Bulk fill performance under a cariogenic challenge provided by *S. mutans* biofilm, since the most

of Bulk Fill composites on the market still remains with BisGMA/TEGDMA on the composition.

In spite of Bulk Fill composites have the purpose to make dentist life easier, they can be subdivided into two groups: the materials that can be exposed to the oral environment (high viscosity), with better mechanical properties; and those that should be used as a base/liner (flowable), in which the manufacturer recommends a capping layer with conventional composite resin and these characteristics are responsible for defining its indications (11). Besides it can be characterized by mechanical tests in which are expected to present variations due to different compositions as different monomers and photoinitiators (12). Thus, due to the great variation presented by material on their composition related with organic and inorganic content, they can be influenced in different ways by biofilm

and also perform in different ways.

Therefore, we hypothesized that biofilm formatted on Bulk Fill can yield lower damaged in composite resins, concerning: 1. roughness and gloss surface; 2. hardness surface; 3. flexural strength. Thus, the aim of this study was to evaluate influence by *S. mutans* biofilm biodegradation on the roughness, hardness, surface gloss and flexural strength of high viscosity Bulk Fill composites.

Material and Methods

Three commercial high viscosity Bulk Fill composites: Filtek Bulk Fill (FBF), Tetric N Ceram Bulk Fill (TNC), X-tra fil (XF) and a conventional Filtek Z350 (FZ) composite were used in this study. The composition, shade, weight (%), lot number and manufacturer of the composites are shown in Box 1.

For all the tests, specimens were randomly distributed in 4 experimental groups (n=10) according to each material, with evaluations accomplished before and after 7 days of biodegradation, regarding the properties of roughness, hardness, surface gloss and flexural strength.

Roughness

Roughness test was carried out on material discs (5 mm in diameter, 2 mm in thickness for all composites) accomplished using a silicone mold (Aquasil LV; Dentsply DeTrey, Maquira, Maringa, PR, Brazil) pressed between two glass slides and covered by polystyrene strips (n=10). The composites were placed in a single increment and the composites Bulk Fill was light-cured for 10 s and Filtek Z350 for 20 s, using a LED source (BluePhase 20i; Ivoclar Vivadent, Schaan, Liechtenstein) with an irradiance of 1,200 mW/cm² as measured using a curing radiometer (Hilux Dental Curing Light Meter; Benilioglu Dental, Demetron, Ankara, Turkey). After storing for 24 h at 37 °C in relative humidity, composite resin discs were polished with abrasive papers in a decreasing sequence of abrasiveness using 1,000-,

Box 1. Material, composition, shade, weight (%), lot number and manufacturer of the composites

Material	Composition	Shade	Weight (%)	Manufacturer #Batch number
Filtek Z350	Silane treated ceramic,		60-80	
	Silane treated silica		1-10	
	UDMA		1-10	
	BisEMA		1-10	
	BisGMA		1.96-5	3M/ESPE, St Paul, MN,USA (90284830233)
	Zirconia ceramic (66402-68-4), surface modified with 3-methacryloxypropyltrimethoxysilane (2530-85- 0),bulk material	A2	<5	
	PEGMA		0.2364	
	TEGDMA		0.99587375 (tipically) 0.037815)	
	Silane treated ceramic		60 - 70	
	Aromatic urethane dimethacrylate		10-20	
Filtek Bulk Fill	Ytterbium dimethacrylate (YbF3)		1-10	
	UDMA		1-10	
	Silane treated silica		1-10	3M ESPE, St. Paul, MN, USA (N867072)
	DDDMA	A2	<5	
	Silane treated zirconia		<5	
	Water		<5	
	Modified methacrylate monomer		<1	
	EDMAB		<0.5	
	Benxotriazol		<0.5	
	Bis-GMA		5-10%	VOCO,Cuxha en,Germany (1702532)
X-tra fil	TEGDMA	Universal	< 2.5%	
	BisGMA		3-<10	IvoclarVivadent, Inc, NY, USA (V19409)
	UDMA		3-<10	
	Yterbium trifluoride	IVA	3-<10	
Tetric N Ceram Bulk Fill	EBPDMA		3-<10	

Bis-EMA= Ethoxylated bisphenol-A-glycidyl methacrylate. Bis-GMA= Bisphenol-A glycidyl methacrylate. UDMA= Urethane dimethacrylate. EBPADMA: ethoxylated bisphenol-A dimethacrylate; TEGDMA= Triethylene glycol dimethacrylate, PEGMA: polyethylene glycol dimethacrylate, DDDMA: 1,12-Dodecane dimethacrylate EDMAB: Ethyl 4-dimethyl aminobenzoate. Information supplied by MSDS available of each product.

J. R. Camassari et al.

1,200-, and 2,000-grit SiC (Norton Abrasives, Vinhedo, SP, Brazil) and finished with diamond pastes 3 μm , 2 μm , 1 μm , and 0.5 μm (Diamond Excel; Buehler) applied with felt discs, according to protocol previously described in the literature (3).

The surface roughness of the specimens was measured using a surface profilometer (Surfcorder SE-1700, Kosaka Laboratory, Tokyo, Japan) with a 0.5 mm radius diamond stylus and 0.01 μm precision, set to 0.25 mm cutoffs and for a total measured length of 1.25 mm at 0.1 mm/s constant speed. Three measurements were made with the diamond stylus passing across the geometric center of the specimens in three different positions on surface by rotating the specimens 90°, 135° and 180° degrees to obtain the arithmetic mean of roughness (Ra- μm).

Knoop Hardness

The specimens (n=10) for each composite were prepared following the same procedures described for the roughness test. After 24 h, the Knoop hardness measurements were conducted on the top surface of the specimens using a microhardness tester (HMV-2; Shimadzu Corp., Tokyo, Japan) with a load of 50 g applied for 15 s. Five indentations were made in the different locations on the top surface and the average value of the five readings was recorded as the Knoop hardness number (KHN) for each specimen.

Gloss Evaluation

Ten specimens for each composite (10 mm in diameter, 2 mm in thickness) were prepared following the same procedures described for the roughness test and Knoop hardness. The gloss-meter measurement principle is based on a light beam incident on the sample surface. The intensity of the reflected light is measured and compared with a reference value. The gloss-meter calibration was made with a highly polished black glass pattern supplied by manufacturer. The surface gloss of the composites resins was evaluated using a 3-angle portable precision gloss-meter (ZGM ; Glossmeter, Zehntner Testing Instruments, Sissach, Switzerland) using a light incidence of 60°. Four measurements were made on each sample and the arithmetic mean of the readings was considered as Gloss values (Gloss Unit - GU). The data obtained was recorded by the software Zehntner Glosstools 1.0.0023.

Flexural Strength

Flexural strength test of the specimens was measured according to the three-point bending carried out with a universal testing machine (Instron, Model 4411; Corona, CA, USA) at a cross-head speed of 0.5 mm/min until specimen fracture. The bar specimens (n=20) were made in dimensions 2 mm x 2 mm x 25 mm for each composite according to

ISO 4049 (13). The specimens were prepared between glass slides and light-cured with three overlapping 60 s using a LED source (BluePhase 20i; Ivoclar Vivadent). After photopolymerization, the bar dimension was measured with a digital caliper (Mitutoyo 293-821; Neuss, Germany) with an accuracy of 0.01 mm. Ten specimens for each composite were stored for 24 h and ten for one week (biodegradations) in dark containers at 37° C. The flexural strength (FS) in MPa was then calculated as: $FS = 3F/l2hb^2$, where F stands for load at fracture (N), l is the span length (20 mm), and b and h are the width and thickness of the specimens in mm, respectively.

Biodegradation

All the specimens subjected to initial tests of roughness, Knoop hardness, surface gloss and flexural strength after biodegradation were sterilized with ethylene oxide in the ACECIL enterprise (Sterilization Center, Campinas, SP, Brazil). *S. mutans* strain UA159 was obtained from a culture of the Department of Microbiology and Immunology, Piracicaba Dental School, State University of Campinas. To prepare the inoculum, *S. mutans* were first grown on *Mitis Salivarius Agar* (Difco Laboratories, Sparks MD, MI, USA) plates at 37 °C for 48 h in an environment supplemented with 10% CO₂. Subsequently, single colonies were inoculated into 5 mL of brain heart infusion (BHI) broth (Difco Laboratories) and incubated at 37 °C for 18 h. Disc specimens were exposed under static conditions to 25 μL of *S. mutans* and the bar shaped specimens to 150 μL inoculum adjusted to an optical density of 0.6 at 550 nm (approximately 8×10^{11} CFU/mL) (6).

After 2 h at room temperature, the non-adhering cells were removed by washing two times with 0.9% NaCl solution (saline). After, a single material disk was placed in each well of polystyrene plates (Nunc multidish 96 well, Sigma, Saint Louis, MO, USA) with 2 mL of sterile, fresh, BHI broth containing 1% of sucrose (wt/vol). Specimens for flexural strength test were stored in test tubes with 4 mL of sterile BHI solution containing 1% of sucrose. The bacterial accumulation occurred at 37 °C in an environment supplemented with 10% CO₂, developing 7 day old biofilm. The medium was renewed at 24 h intervals. At the end of the experimental period, specimens were ultrasonically washed for 10 min and the final tests for surface roughness, Knoop hardness, surface gloss and flexural strength were performed as described for the initial period. Representative samples of each material (n=3), before and after biodegradation, were sputter-coated with gold under vacuum (Balzers-SCD 050 sputter coater, Balzers, Liechtenstein) and examined using scanning electron microscope (Model JEOL JSM 5600 LV, Tokyo, Japan) operating at 1,000 x magnification. X-ray (EDX)

spectrometry, coupled to a scanning electron microscope (JEOL, JSM-5600LV, Tokyo, Japan) was performed to identify the elemental composition of the surface filler particles. Each spectrum was acquired for 100 s (voltage 15 kV, dead time 20% to 25%, working distance 20 mm). Images showing the identified chemical elements and their relative concentration were obtained from three different analyses of each material at three different locations on a stub.

Statistical Analysis

Data were tested for normality (Kolmogorov-Smirnov). Data for roughness, Knoop hardness and surface gloss were analyzed with two-way ANOVA. Multiple comparisons were performed using the Tukey post-hoc test ($\alpha = 0.05$). Data of the flexural strength were analyzed with two-way ANOVA analysis of variance and Tukey post-hoc test ($\alpha = 0.05$). All test was conducted considering the significance of 5%, using SPSS software 22.

Results

Table 1 shows the surface roughness of the composites before and after the biodegradation. Significant differences in roughness for composites ($p < 0.000$) and biodegradation ($p < 0.000$) were detected. The interaction between the composites and biodegradation was significant ($p = 0.003$).

Table 1. Means of roughness \pm Standard deviation (μm), before and after biodegradation by *S. mutans* for the composites

Composites	Biodegradation	
	Before	After
Filtek Z350	0.0944 \pm 0.037 Bb	0.1443 \pm 0.044 Ac
Filtek Bulk Fill	0.0787 \pm 0.007 Bb	0.1673 \pm 0.066 Ab
X-tra fil	0.1251 \pm 0.042 Ba	0.3100 \pm 0.118 Aa
Tetric N Ceram Bulk Fill	0.0861 \pm 0.010 Bb	0.1766 \pm 0.048 Ab

Values followed by the different lower-case superscript within the same column and upper-case superscript in the same row are statistically different ($\alpha = 5\%$).

Table 3. Means of Knoop hardness \pm Standard Deviation (KHN), before and after biodegradation by *S. mutans* for the composites

Composites	Biodegradation	
	Before	After
Filtek Z350	97.0 \pm 23.8 Ab	77.2 \pm 12.0 Ba
Filtek Bulk Fill	86.6 \pm 12.6 Ab	66.7 \pm 10.4 Ba
X-tra fil	151.7 \pm 27.8 Ab	106.0 \pm 27.2 Bb
Tetric N Ceram Bulk Fill	62.2 \pm 5.6 Ac	51.8 \pm 8.6 Bb

Values followed by the different lower-case superscript within the same column and upper-case superscript in the same row are statistically different ($\alpha = 5\%$).

Surface roughness increased for all materials after *S. mutans* biodegradation. Comparing different composites before and after biodegradation, the highest roughness values was obtained for the composite XF ($p < 0.05$).

Table 2 shows the gloss results before and after biodegradation. There was significant difference between the materials ($p = 0.010$) and biodegradation ($p = 0.005$). However, there was no interaction between the material x biodegradation factors ($p = 0.845$). It was observed that FZ and FBF showed the highest gloss values before and after the biodegradation in relation others materials ($p < 0.05$). TNC and XF showed similar lower gloss values. It was observed reduction of gloss values for all the composites after biodegradation ($p < 0.05$).

Table 3 shows the Knoop hardness results of the composites before and after biodegradation. There was significant difference between the materials ($p < 0.000$) and biodegradation ($p < 0.000$). The interaction between the material and biodegradation factors was significant ($p = 0.024$). The surface hardness values for all materials decreased significantly after biodegradation ($p < 0.05$). It was observed that before and after biodegradation, the XF composite presented the highest values for hardness when compared to other materials ($p < 0.05$) and the lowest value was shown by TNC, while FBF and FZ presented

Table 2. Means of gloss \pm Standard Deviation (GU), before and after biodegradation by *S. mutans* for the composites

Composites	Biodegradation	
	Before	After
Filtek Z350	71.7 \pm 11.2 Aa	62.0 \pm 9.6 Ba
Filtek Bulk Fill	69.0 \pm 9.9 Aa	64.6 \pm 8.4 Ba
X-tra fil	58.5 \pm 13.4 Ab	53.5 \pm 11.6 Bb
Tetric N Ceram Bulk Fill	61.4 \pm 5.7 Ab	53.3 \pm 9.2 Bb

Values followed by the different lower-case superscript within the same column and upper-case superscript in the same row are statistically different ($\alpha = 5\%$).

Table 4. Means of flexural strength \pm Standard Deviation (MPa), before and after biodegradation by *S. mutans* for the composites

Composites	Biodegradation	
	Before	After
Filtek Z350	127.6 \pm 15.3 Aa	99.0 \pm 12.1 Bbc
Filtek Bulk Fill	105.6 \pm 14.8 Abc	104.2 \pm 15.8 ABab
X-tra fil	117.7 \pm 5.6 Aab	117.7 \pm 16.1 Aa
Tetric N Ceram Bulk Fill	86.9 \pm 9.0 Ac	81.9 \pm 5.4 Ac

Values followed by the different lower-case superscript within the same column and upper-case superscript in the same row are statistically different ($\alpha = 5\%$).

intermediate values.

Table 4 shows the results of the flexural strength of the composites before and after the biodegradation by *S. mutans*. There was a significant difference between the materials ($p < 0.000$) and for biodegradation factors ($p = 0.003$). The interaction between material x biodegradation was significant ($p = 0.004$). The composite FZ showed the highest flexural strength despite it showed the significant decrease on the values caused by biodegradation ($p < 0.05$). There was not observed significant difference between FBF and XF on flexural strength values, as well as between FBF and TNC. After biodegradation, the composite

XF showed the highest flexural strength values compared to TNC and FZ ($p < 0.05$). There was no significant difference between FZ and FBF, and between FBF and XF, as well as between FZ and TNC concerning flexural strength values ($p > 0.05$). When flexural strength was compared before and after biodegradation, there was a significant reduction in the flexural strength value only for the FZ composite ($p < 0.05$).

Characterization and filler content images are illustrated on Figure 1. The SEM micrograph showed several sizes and shapes fillers, but mostly round, and some cluster formations (black star) (Fig. 1A and 1C). The elemental

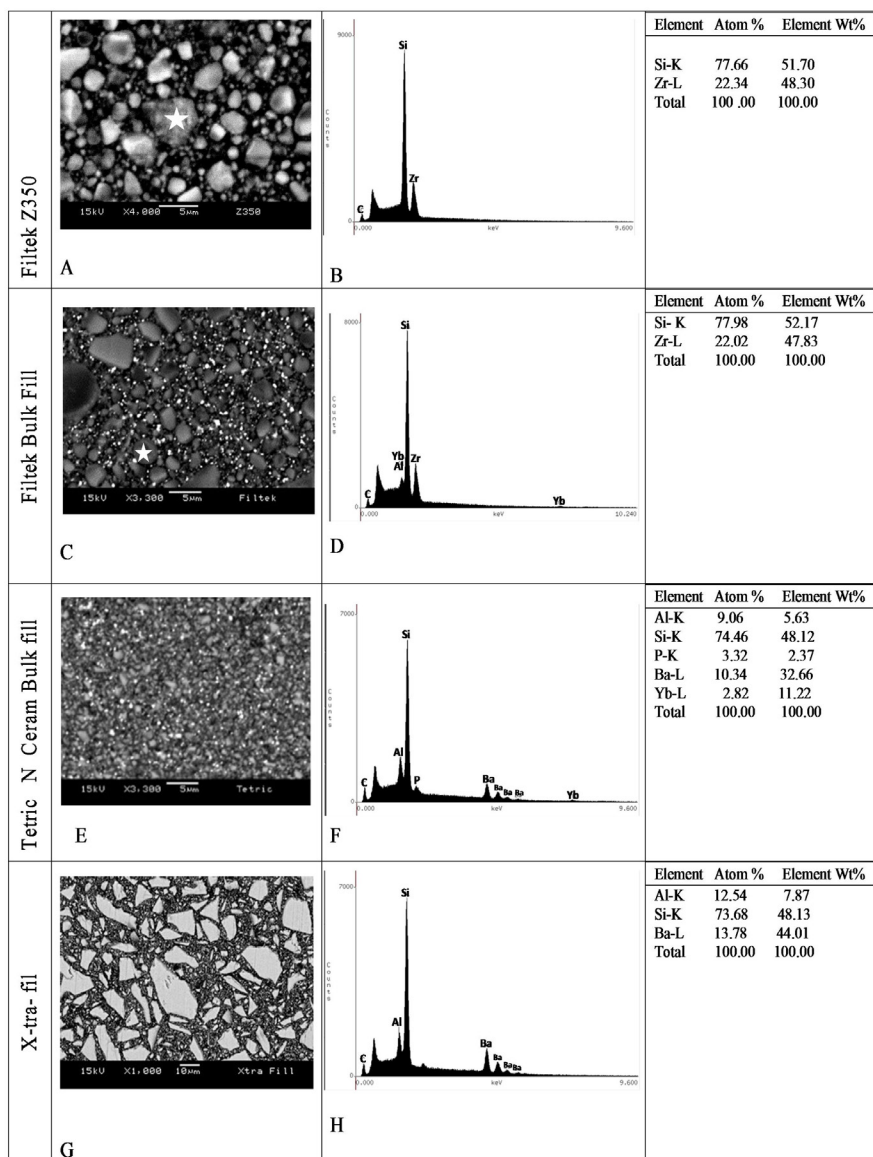


Figure 1. Representative scanning electron microscopy filler characterization of all composites before biodegradation in Back Scattering, showing the size, filler morphology and content represented by EDS. A. several sizes and shapes fillers, but mostly round, and some cluster formations (white star); B. $> 5 \mu\text{m}$ fillers (white star) mixed with several sizes and shapes, including nanofillers; C nanofillers mixed with round and irregular shapes; D. Irregular fillers with different sizes but mostly $> 10 \mu\text{m}$. Magnification from 1000x to 4000x.

composition of FZ revealed the presence Carbonum, Silica and Zirconium (Fig. 1B). FBF had a morphology similar to FZ; also the value of fillers >5 µm is estimated from SEM (Fig. 1C). The inorganic elements in FBF were found to include Carbonum, Aluminium, Yterbium, Silica and Zirconium (Fig. 1D). The TNC images showed the presence of spherical and irregular nano-sized fillers (Fig. 1E) and EDS revealed Carbonum, Aluminium, Silica, Phosphorum, and Barium (Fig. 1F). The particles morphology for XF were constituted by irregular fillers, basically, but, mostly higher

than 10 µm (Fig. 1G). The filler content was similar to TNC, except by absence of Phosphorum (Fig. 1H).

Figure 2 shows representative scanning electron microscopy (SEM) images from composite resin surfaces before and after degradation by *S. mutans* biofilm. It can be observed the shape and distribution of the fillers from each composite resin studied. After biodegradation, it is possible observe a smoother surface with uniform distribution of filler particles for the FZ composite (Fig. 2E). For XF composite (Fig. 2H), is possible to notice the presence of

J. R. Camassari et al.

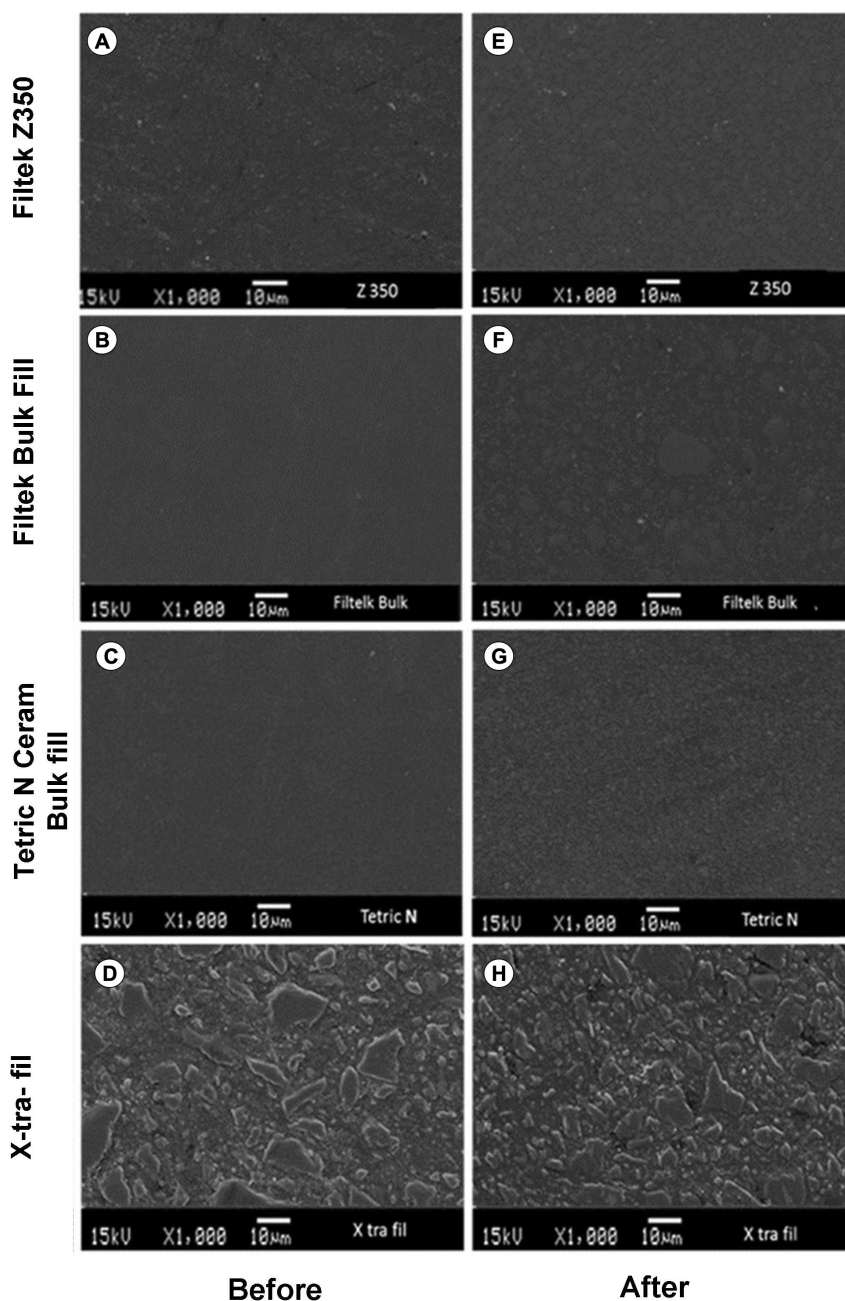


Figure 2. Representative scanning electron microscopy before and after degradation respectively of Filtek Z350 (A, E), Filtek Bulk (B, F), Tetric N-Ceram Bulk Fill (C, G) and Fill X-tra fil (D, H). Magnification of 1000x.

larger sized particles with irregular shape and distribution. TNC (Fig. 2G) presented a slightly corroded surface and on the surface of FBF composite (Fig. 2F) may be observed the exposure of intermediate sized filler particles.

Discussion

The first hypothesis that the biofilm provide lower damage on roughness and gloss surface of Bulk Fill composite resins was rejected, since those properties were influenced not only by biofilm, but also by material composition.

In order to obtain aesthetics and longevity in composite resin restorations, surface smoothness is required considering that surfaces with roughness greater than 0.2 μm allow an increase in biofilm accumulation (14). According to the results obtained in this study, before biodegradation the composite XF showed the highest roughness when compared to other composites. Previous study showed that this result is related to highest percentage in volume (70.1%) and size (2–3 μm) of the filler (15). Furthermore, Marghalani has shown that the shape of the filler particles also influenced the surface roughness (16), suggesting that irregular fillers (Fig. 1G and 2D), such as XF composite, promote rougher surfaces. The composite FZ, FBF and TNC showed similar roughness values before biodegradation (Table 1, Fig. 2A, 2B and 2C). As it can be seen in Box 1, these composites have in the composition filler particles with reduced size and similar or lower percentage in volume when compared to XF composite. The filler size allows better polishing and, consequently, higher surface smoothness (16,17).

After biodegradation, there was an increase of roughness for all the composites (Table 1 and Fig. 2). Although this increase is due to the production of acids, which are able to promote surface changes (5,7), others factors, as filler size and monomer composition, may have influenced the results. According to Montanaro et al. (17), the adhesion of *S. mutans* to surface of the composites is related to size, shape and distribution of the charge particles, besides the composition of the organic matrix. As it can be seen in Figures 1G and 2D, the XF composite showed irregular and bigger filler particles, that makes surface rougher, allowing greater adhesion of *S. mutans*, reflecting on higher roughness value, even after polishing. Adversely, in the conventional composite FZ there are fillers combination involving silica nanofillers filling the spaces left by zirconia-silica nanoclusters. The reduced size and homogeneous distribution of the filler particles in this composite (Fig. 2A) resulted in lower *S. mutans* adhesion, similar to previous study (17). It is also known that in this nanocomposite there is lower interstitial space among the filler particles and, consequently, less exposure

of the organic matrix (18), making it more resistant to biodegradation. Therefore, these factors are responsible for the lower surface roughness of FZ after biodegradation. TNC and FBF composites showed intermediaries and did not differ statistically in roughness after biodegradation. This result can be attributed to similarity of size and volume percentage of the filler particles of these materials (16).

It is claimed that the surface gloss of the composites shows an inverse relation with roughness (19), corroborating with the results obtained in the current study. It can be observed in this study that after biodegradation, besides the increase of surface roughness, all composites presented reduction in the brightness values. This result is certainly because of the surface changes that occurred in the materials exposed to the cariogenic biofilm (6,7), affecting the incidence and reflection of the light, promoting reduction of the surface gloss of the composites (20). It can be seen in Box 1, that composite XF present a higher amount of TEGDMA than composite FZ, about 10 times and other composites does not show that monomer. Consequently, changes on the organic matrix and the interaction between matrix and filler particles may promote significant reduction of the surface gloss of composites (21). However, when the materials were compared before and after biodegradation may be observed that higher gloss values were obtained for composites FZ and FBF. Small filler particles of those composites (Fig. 1A and 1C) caused a decrease in diffuse reflection and a high refractive index resulting in higher gloss values compared to the others composites (11,22). Nevertheless, lower gloss values was obtained by composites XF and TNC, which can be explained by the larger size (composite XF) of their particles (Fig. 1G) and irregular shape (Fig. 1E and 1G) that caused a lower homogeneity of the matrix-filler complex resulting in low light reflection (20).

Surface hardness is an important property that is directly related to mechanical performance and depends on the degree of polymerization of the material, the size, weight and volume of the inorganic fillers, as well as the composition of the composite matrix (9). Composite XF presented higher Knoop hardness value, which may be attributed to higher size and percentage of filler in volume, being one of the factors responsible for increasing the performance of the mechanical properties (23). According to Ilie et al. (10), increasing the size of the inorganic fillers reduces the matrix amount incorporated into the material and, consequently, the matrix-filler interface. Thus, light scattering is reduced during photo-activation due to higher absorption, increasing of the degree of conversion and resulting in higher hardness values.

However, lower Knoop hardness value was observed for the composite TNC. Even though the photoinitiator is part

of the composition, which according to manufacturer would provide increase at the polymerization depth, it also contains prepolymers that may be related to lower Knoop hardness values because prepolymers are calculated together with the volume percentage of the inorganic matrix (10). The composites FZ and FBF presented intermediaries Knoop hardness values and this may be attributed to the similar size and shape of the inorganic fillers (22).

Biofilm degradation by *S. mutans* caused a significant decrease in hardness of all materials. There is a great possibility that the functional groups -OH and -CHOO of the acids produced by the bacteria form hydrogen bonds with the polar side of the methacrylate monomer of the composites, causing greater water absorption and softening of the resin matrix. This fact induces tension at the matrix-silane-filler interface separating the inorganic fillers from the organic matrix, promoting hardness reduction (24).

Flexural strength test was performed according to ISO 4049 standard. Before biodegradation, Filtek Z350 composite presented higher value of flexural strength when compared to the composites TNC and FBF. The flexural strength values suggest that higher inorganic particle content does not necessarily result in higher flexural strength values. In addition to the filler content, other factors such as tension between fillers and resin matrix, as well as the bond between the components and the composition of the organic matrix (23) may be relevant on the flexural strength of these materials. The best results in mechanical properties of FZ may be related to a higher degree of monomeric conversion already confirmed by another study (12). However, the composite TNC showed lower value when compared to composites XF and FZ. As previously discussed, despite the incorporation of the Ivocerim photoinitiator as an attempt to improve the polymerization depth, the presence of prepolymers in the composite may have negatively influenced on the mechanical properties, reflecting the lower value of flexural strength for this material (25). In addition, the shorter photoactivation time indicated by the manufacturer may not have been enough to obtain adequate polymerization.

When comparing the composites after biodegradation, it may be observed that the composite XF presented a higher value of flexural strength when compared to the composites TNC and FBF. Probably, this result would be related to the larger size and volume of the filler particles (Fig. 1G) (25). Despite the differences found in flexural strength values when comparing the different composites, the degradation by *S. mutans* biofilm was able to reduce only the resistance of the FZ composite. This result may be related to different compositions of the organic matrices of these materials. In addition, the composite FZ showed highest amounts of zirconia-silica clusters (Fig. 1B), responsible for increasing

the water absorption by the material due to the higher volume of silane, which makes it more susceptible to hydrolytic degradation (24)

In summary, the results showed increased roughness and decreased hardness and gloss of all composites evaluated. No effect on the flexural strength occurred for the Bulk Fill composites. Reduction of flexural strength occurred only for the conventional FZ composite. The biodegradation by *S. mutans* negatively affected the mechanical and surface properties of the materials. Care should be taken by clinicians during the selection of the restorative material and the knowledge of the patient about the importance of the continuous use of oral hygiene techniques is essential for maintaining aesthetics and longevity of composite resin restorations. Future studies should be developed to investigate other possible factors such as mechanical brushing and different light-curing source.

Resumo

O objetivo deste estudo foi avaliar a influência do biofilme de *S. mutans* na rugosidade (Ra), brilho (GU), dureza superficial (KHN) e resistência à flexão (FS) de compósitos de Bulk Fill de alta viscosidade. Foram utilizados Filtek Bulk Fill (FBF), Tetric N Ceram Bulk Fill (TNC), X-tra fil Bulk Fill (XF) e Filtek Z350 (FZ). Dez discos de cada compósito foram preparados para Ra, KHN e GU e 20 barras para o FS. Após 24 h, os espécimes foram polidos e as análises iniciais realizadas. As amostras foram esterilizadas e submetidas a biodegradação por 7 dias e as análises finais foram realizadas. Amostras representativas de cada grupo foram avaliadas no Microscópio Eletrônico de Varredura. Os dados foram submetidos à ANOVA dois fatores e teste de Tukey. XF apresentou os maiores valores ($p < 0,05$) de Ra antes e após a biodegradação (0,1251; 0,00,3100) e FZ (0,1443) os menores após a biodegradação ($p < 0,05$). Os maiores valores de GU ($p < 0,05$) foram observados para FZ (71,7; 62) e FBF (69,0; 64,6), e os mais baixos ($p < 0,05$) para TNC (61,4; 53,3) e XF (58,5; 53,5), ambos antes e depois da biodegradação. Para KHN, os valores mais altos foram obtidos por XF (151,7; 106), e os ($p < 0,05$) valores mais baixos para TNC (62,2; 51,8), antes e depois da biodegradação. Os maiores valores ($p < 0,05$) de FS foram observados para a FZ (127,6) e os menores ($p < 0,05$) para a TNC (86,9); após biodegradação, o XF (117,7) apresentou os maiores valores ($p < 0,05$) em comparação ao TNC e FZ. Em conclusão, a biodegradação aumentou o Ra e diminuiu a GU e a KHN para todos. Em relação à FS, a degradação promoveu uma diminuição significativa apenas para a FZ.

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