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Effects of cellulose fibers on the physical and chemical properties of glass ionomer dental restorative materials

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ABSTRACT

A dental glass ionomer cement (GIC) was modified with cellulosic fibers. The microstructural analysis and physicochemical properties were evaluated in four groups: GIC (control) and GIC modified with different concentrations of fibers, GICMF1, GICMF2 and GICMF3. Within clinically acceptable limits, composites showed capacity of water absorption and solubility in water similar to GIC and no signs of disintegration were observed. GICMF2 provided greater resistance to compression, wear and adhesion, however it had no effect on the diametral tensile strength. Morphological and chemical element analyses of GICMF2 showed the formation of a new and stable composite with interaction between fiber/ionomer matrix/load particles.

1. Introduction

The widespread use of polyalcenoate glass cements called glass ionomer cements as restorative dental material is related to its bond to tooth structure, coefficient of linear thermal expansion similar to dentin, biocompatibility and storage and fluoride releasing capacity [1–4]. Due to their chemical characteristics and biocompatibility, indications for their use have been extended to other fields as well. For example, glass-ionomer cements are widely used in medicine, primarily in otology, reconstructive surgery, orthopedics [5] and also as a structural material (scaffold) for bone formation [6,7].

Nevertheless, glass-ionomer cements also have some mechanical and clinical limitations as dental restorative materials. In addition to having a poor esthetic appearance due to their limited translucency, and technique sensitivity, they have a short working time and prolonged setting period, in which first 24 h are critical, making this material susceptible to hygroscopic change in the medium and solubility [8]. Thus, immediate surface protection of the glass ionomer cement restoration with a waterproofing material is indicated [9–11].

The chemical bonding capacity of glass ionomer cement to dental structures is highlighted. There is a chemical interaction between the carboxylic groups of polyacids, which are chelating agents within the cement, and calcium ions in the tooth enamel and dentin [12]. Although this is a positive feature of the material, it is known that its bond strength is still considered low [13]. Another feature of these cements is the deficiency of their mechanical integrity and their ability to withstand fracture loads [8]. Many studies have reported the degree of deformation of glass ionomer cements, by testing the compressive, diametral tensile, bond strength and wear strength [14–17].

Successive changes have been made in the conventional glass ionomer cement, in order to increase its mechanical strength. Thus, several materials have emerged with different compositions such as glass ionomer cements reinforced with metal, modified with resin, and finally the high-viscosity ionomer cements and those with incorporation of nanoparticles, all to meet individual clinical needs and improve their physicochemical properties [8,18–20].

For many years there have been attempts to incorporate fibers into the composition of these materials as agents to reinforce their physical structure [21,22]. In recent decades there has been rapid development of natural fiber-reinforced composites, giving them better mechanical strength and an increase in elasticity modulus [22–24].

The use of these fibers in composites requires compatibility between the fibers and matrix, the incorporation of a relatively large amount of fibers and wettability capacity, factors that increase the load transfer to them [23]. Cellulosic fibers meet these needs, because although they have interesting features for use, such as low cost, low density, specific strength and high elasticity modulus, they have a high number of fibers per gram, which provide resistance to flatness and rigidity [22,24]. Moreover, they

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are not abrasive or toxic, can be easily modified by chemical agents and produced with different levels of whiteness and a very low content of impurities, they are abundant and come from renewable sources [25].

Numerous studies with cellulose fibers added to composites have shown proven results of superior strength. There was higher flexural strength, higher tensile strength and higher elasticity modulus of composites modified with these fibers [21,22]. However, no study has been reported in the literature involving the use of cellulosic fibers in dental materials.

The aim of this study was to modify a conventional restorative glass ionomer cement with cellulosic fibers, assess its physico-chemical properties and characterize the composite and its precursors by light microscopy, scanning electron microscopy and energy dispersive X-ray spectroscopy.

2. Experimental procedure

2.1. Composite preparation

The cellulose fibers were obtained from eucalyptus wood and individualized with sodium hydroxide and sodium sulfide (Kraft cooking process), according to the procedures reported by Ferreira et al. [26]. They were then bleached by a sequence of three stages (OD (PO)), composed of oxygen delignification (O), chlorine dioxide delignification (D) and alkaline extraction with pressurized hydrogen peroxide (PO). Fibers were obtained with a desired final brightness of around 80% ISO [27], a color very close to the natural color of teeth. The material was stored in plastic containers at an approximate temperature of 6 °C to avoid fungal proliferation.

Fibers were weighed on analytical balance and then subjected to the drying process in an oven at 37 °C (±1 °C) for 24 h.

Increasing concentrations of fibers were added to the commercially available, conventional restorative glass ionomer cement (GIC), during cement handling in accordance with the recommendations of the manufacturer. Three different types of glass ionomer cement composites modified with fibers (GICMF) were prepared according to the concentration of fibers agglutinated in the cement. Thus, four study groups were obtained: group 1 (G1), GIC, control group; group 2 (G2), GICMF1; group 3 (G3), GICMF2; group 4 (G4), GICMF3.

2.2. Evaluation of syneresis and imbibition

Test samples (n = 12) of all groups were fabricated in a metal matrix measuring 5 mm in diameter and 3 mm deep, resting on a glass plate. In G1 the cement was inserted into the matrix under pressure, through a special syringe (Centrix, DFL Ind., SP, Brazil) and in G2, G3 and G4 cement was inserted into the matrix with a putty knife and compacted with the aid of an amalgam condenser. After the matrix was completely filled, a polyester strip was pressed onto it, under a 500 g weight, until it reaches its setting time in order to obtain adequate flow and surface smoothness. The groups of test samples were divided randomly into two groups (n = 6), a group using surface protection with dental cavity varnish (A) and the other without protection (B). All samples were stored in distilled water for 24 h.

Test samples were dried rapidly with absorbent paper and one of the diameter dimensions, with and without protective varnish, was marked with a copy pencil to evaluate the syneresis. Initial measures (IM) were obtained with a caliper and after this, the test samples were kept at room temperature with controlled humidity. For evaluation of imbibition 12 h after drying, test samples were weighed (IW) in an analytical balance and immersed in 50 mL distilled water in closed containers at 37 °C (±1 °C).

Test samples were measured (FM) and weighted (FW) again at intervals of 24 h and 7, 14 and 30 days after which it was possible to assess their loss and gain of water, respectively, by the difference between initial and final measurements (FM–IM) and the difference between initial and final weights (IW–FW), respectively.

2.3. Evaluation of solubility and disintegration

Test samples (n = 12) of all groups were made for the solubility test in the manner described above. After the cement had set, each of the test samples was weighed on a precision scale and immersed in 50 mL distilled water in individual and identified containers in an oven at 37 °C (±1 °C). After 7 days, the test samples were washed thoroughly with distilled water to remove any surface residues and the excess moisture was removed by drying for 12 h. Subsequently, they were weighed again and the solubility value, obtained from the difference between the initial (IW) and final weighing (FW) was calculated.

The disintegration of test sample surfaces was assessed by observation under a stereomicroscope (Ken-A-Vision, USA).

2.4. Compressive strength and diametral tensile strength tests

The test samples (n = 24) of G1, G2 and G3 were made using a Teflon matrix measuring 4 mm in diameter and 8 mm long, in the manner described for evaluating the syneresis and imbibition. After 7 days of storage in distilled water at 37 °C (±1 °C), the test samples (n = 12) were tested for compressive strength in a universal testing machine (DL2000, EMIC, SP, Brazil) with load cell of 200 kgf at 1 mm/min, with their long axes in a vertical position until fracture. To test their diametral tensile strength, the test samples (n = 12) were subjected to the same load cell, but at a speed of 0.5 mm/min and their long axes in the horizontal position.

2.5. Wear resistance test

The test samples (n = 12) of G1 and G3 were prepared as before, and immediately after this they were stored in distilled water at 37 °C (±1 °C) for 14 days. After drying for 12 h, test samples were weighed on a precision scale and each one was adapted to the center of an acrylic resin base with central perforation of the same diameter as the test specimen. Six bases with test samples were adapted to an appropriate device, which was placed on 800-granulation sandpaper on a metallographic sanding machine (Model PLFDV, Fortel, SP, Brazil) and submitted to an abrasive wear test cycle. In every cycle a 100 g load was used at 100 rpm for 30 min under constant water irrigation. The sandpaper used was replaced by a new one after every six test specimens had been submitted to the abrasion test.

After abrasive wear, the test samples were washed in distilled water for 3 min under slow agitation. Again, test samples were dried for 12 h and weighed in order to assess the amount of structure lost by mechanical action.

2.6. Bond strength test

The design of this study was submitted to and approved by the Research Ethics Committee of the Federal University of Vales do Jequitinhonha e Mucuri (CEP/UFVJM) and filed under Registration Number 044/09.

The test samples were prepared from the selection of ten intact human premolar teeth, freshly extracted for orthodontic reasons, free of any changes that could impair bonding. The teeth were cleaned and initially sectioned transversely in a metallographic cutter (ELSAW, Elquip, SP, Brazil) and the roots were discarded. Each tooth crown was sectioned along its long axis, making 20 hemi-sections, which were embedded in ¾ PVC tubes with

polyester resin. After polymerization, each hemi-section was slightly polished in a metallographic polisher under cooling, using 600 granulation sandpaper to expose and simulate a surface layer of dentin. After this, the surfaces were washed and the specimens were stored in distilled water at 37 °C (±1 °C). The hemi-sections were distributed randomly into two groups ($n = 10$), in which restorations in dentin, measuring 3 mm in diameter and 2 mm deep were simulated, using GIC (G1) and GICMF2 (G3).

The dentin surface was etched with 11.5% polyacrylic acid and after being washed and dried with absorbent paper, the restoration was performed. Materials were handled in accordance with the procedures already mentioned. All test samples were again stored in distilled water at 37 °C (±1 °C). After 7 days, test samples were tested for microshear bond strength on a universal testing machine using load of 200 kgf at 0.5 mm/min. Thus, a shear stress was developed at the tooth structure base/ionomer cement interface until the adhesive bond was broken.

2.7. Statistical analyses

After testing for normality (Shapiro–Wilk) and homogeneity of variance (Levene Statistical test), the results of mechanical properties were submitted to specific statistical tests for analysis of the differences between groups and individuals. A 95% significance level ($p < 0.05$) was used for all analyses in the SPSS software program for Windows (Statistical Package for Social Sciences) version 13.0 (SPSS Inc., USA).

2.8. Light microscopy (LM)

Cellulose fibers, as presented in nature, were stained with Astra Blue, placed on a glass plate covered with a glass coverslip and photographed. The processed fibers were distributed on a histological slide, onto which transparent liquid resin was dripped, and then covered with a glass coverslip. GIC powder was dispersed in a solution composed of 50% ethyl alcohol 96° and 50% distilled water and dripped onto a glass slide and covered with a coverslip. The morphology of fibers and powder was observed under a stereomicroscope coupled to an image capture system Motic 1000 (MOTIC) and also observed under an inverted light microscope (Telaval, Zeiss), where they were photographed. The morphology of significant samples of composites of all groups was observed and photographed under the stereomicroscope.

2.9. Scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDS)

Fibers were immersed in distilled water for 30 min under gentle agitation, and treated with increasing concentrations of acetone solutions. Subsequently, they were dehydrated, identified and dried at room temperature for approximately 12 h. Samples of the dried fibers were treated with carbon and significant samples of composites were covered with a thin layer of gold–palladium (~15 nm) for a 2-min deposition time, 1×10^{-1} bar vacuum and 25 mA deposition current using a sputter coater model 550× (Electron Microscopy Sciences) to enable the transmission of electrons on these surfaces. Samples were analyzed by scanning electron microscopy (model 1430 VP, Leo Electron Microscopy Ltd., England and model CS-3500, Shimadzu, Japan) and energy dispersive X-ray spectroscopy (EDS) (CS3200, Oxford, England) using a 15–20 keV electron beam. Results were determined by surface analysis of three different areas of each sample for detailed analysis of their morphology and organization, as well as to obtain three-dimensional images for viewing.

3. Results

The composites GICMF1 (G2) and GICMF2 (G3) were shown to be easy to handle, with minimal reduction in their working time and wettability aspect, observed by the surface brightness of the cementitious mass, when compared with GIC (G1). The ease of insertion and compression capability of the GICMF1 and GICMF2 matrices showed the best consistency of these cements. The composite GICMF3 (G4) showed high viscosity and a considerably lower working time, making it difficult to incorporate the cement into the matrix. After the setting time, the composites developed were stable, with complete aggregation of the fibers in the cement mass.

3.1. Syneresis and imbibition

The analysis of variance with a fixed criterion showed no statistically significant difference between G2, G3 and G4 compared with G1 after time intervals of 24 h and 7, 14 and 30 days for syneresis (Fig. 1) and imbibition (Fig. 2) in the test samples without protective varnish. The composite G2, G3 and G4 showed no statistically significant difference within groups.

The evaluation of syneresis in the test samples with varnish protection showed no statistically significant difference within groups after time intervals of 24 h and after 7 days, but statistically significant differences after 14 and 30 days. The Tukey test showed that G3 had higher syneresis compared with G1 and showed no difference in comparison with the other groups, which showed no statistically significant difference among them after 14 days. After 30 days, there was statistically significant difference in G1 compared with G2 and G3, which showed no difference between them. G3 showed no statistically significant difference in comparison with the other groups (Fig. 1). In the evaluation of imbibition, it was observed that after time intervals of 24 h and 7,

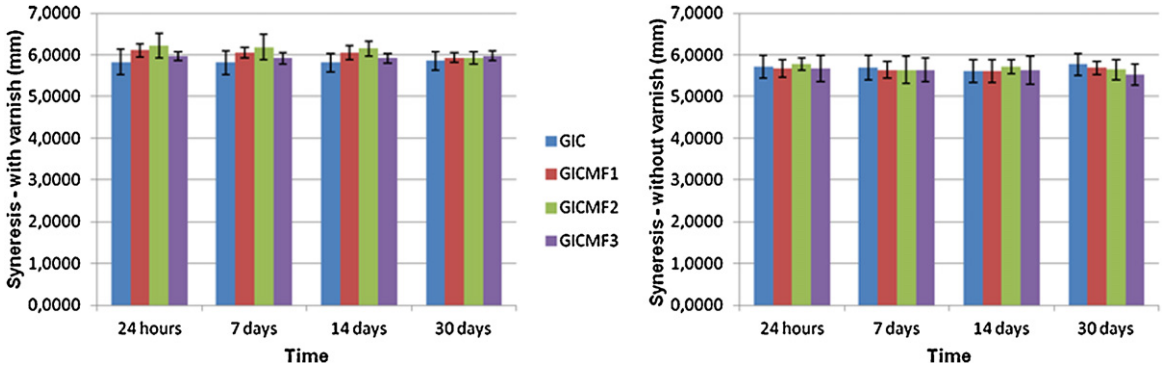

Fig. 1. Means and standard deviations for the syneresis of sample groups with and without protective varnish.

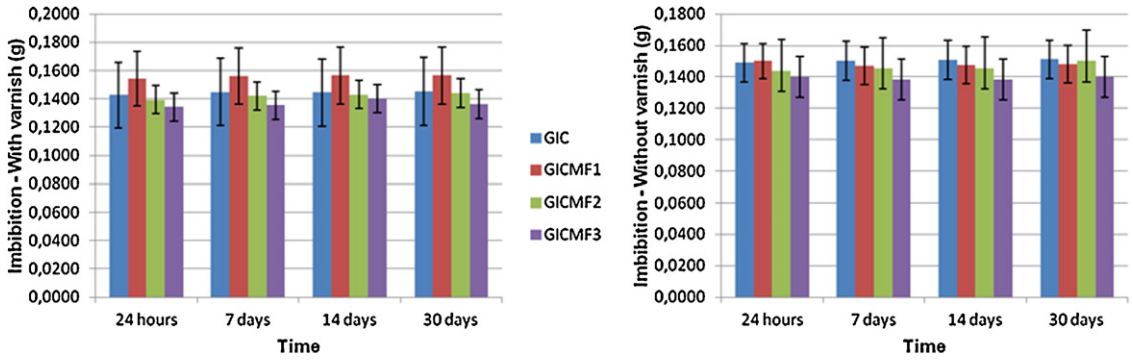

Fig. 2. Means and standard deviations for the imbibition of sample groups with and without protective varnish and significance of the analysis of variance.

14 and 30 days there was no statistically significant difference within the groups (Fig. 2).

3.2. Solubility and disintegration

The composite groups showed statistically significant difference ($p < 0.010$) between each other when solubility was assessed by means of the analysis of variance. The Tukey test showed higher solubility values for composites modified with fibers in G4 (0.78%), G2 (0.58%) and G3 (0.51%) when compared with G1 (0.41%). The composites in G2, G3 and G4 showed no significant signs of disintegration, when compared with G1 after cursory examination of the test samples under stereomicroscope (Fig. 3).

3.3. Mechanical strength

The analysis of variance showed that there was statistically significant difference ($p < 0.001$) for compressive strength, and the Tukey test, that this property was higher in the composites in G2 and G3 compared with those in G1. There was no statistically significant difference ($p = 0.340$) between the groups for diametral tensile strength. According to the Student's *t*-test for independent samples, there was statistically significant difference between groups G1 and G3 for wear resistance ($p = 0.001$) and bond strength ($p = 0.047$) (Table 1).

3.4. ML

The natural cellulose fibers were shown to be a tangle of fibers with a fine and rigid structure, approximately 1 mm in size (Fig. 4a). After processing, they presented as an irregular cluster with a low weight and whitish color (Fig. 4b). When observed under the inverted light microscope, the fibers were shown to be grooved and hollow-looking (Fig. 4c). The GIC powder was shown to be dense and fine grained, with a smooth texture and whitish in color. The presence of transparent, heterogeneous-sized particles was observed, suggestive of glass particles (Fig. 4d–f).

GIC (Fig. 3a) showed incorporation of surface bubbles after powder/liquid agglutination. The glass ionomer cement

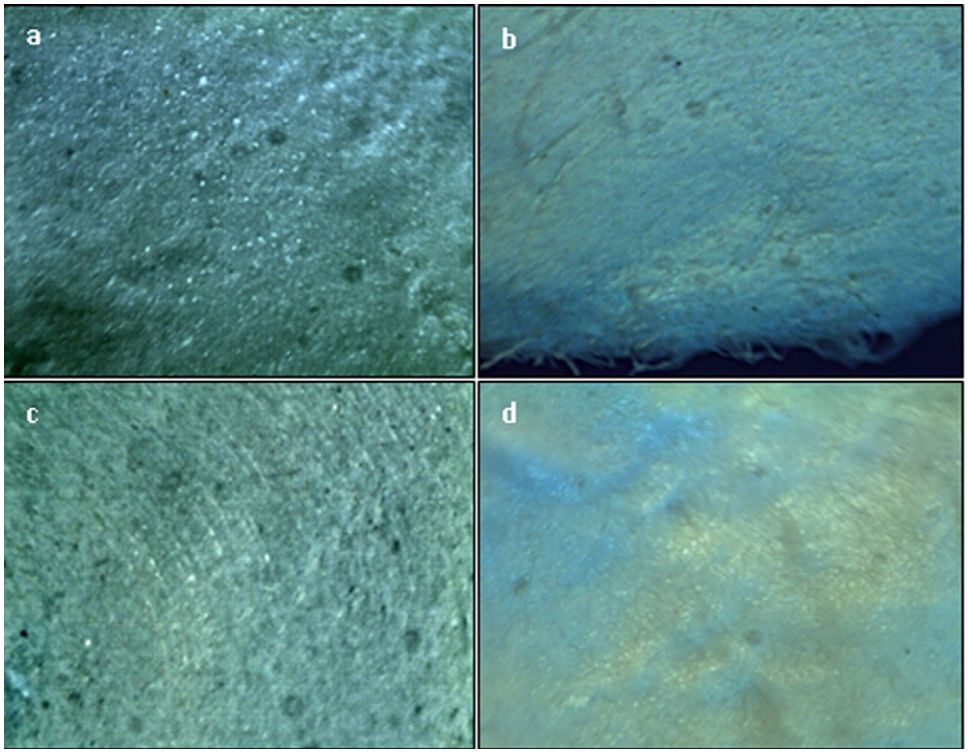

Fig. 3. Photographs of the surface of GIC (a), GICMF1 (b), GICMF2 (c) and GICMF3 (d) under a stereoscope microscope at 45× magnification.

Table 1
Means and standard deviations for compressive strength, diametral tensile strength, wear and adhesion of sample groups and statistical tests.

Group ($n = 10$)	Compressive strength (MPa)		Diametral tensile strength (MPa)	Wear resistance weight loss (g)	Bond strength (MPa)
	Mean (SD)	$p < 0.001$* Tukey**	$p = 0.340$*	$p < 0.001$***	$p = 0.047$***
			Mean (SD)	Mean (SD)	Mean (SD)
GIC	49.150 (46.000)	A	11.700 (14.100)	0.081 (0.002)	0.008 (0.236)
GICMF1	114.690 (12.700)	B	13.570 (20.100)	–	–
GICMF2	62.570 (18.100)	A	13.000 (5.520)	0.053 (0.012)	0.055 (0.948)

* ANOVA.
** Same letters indicate no statistically significant difference.
*** Student's *t*-test.

composites modified with fibers were more compact and homogeneous (Fig. 3b–d).

3.5. SEM/EDS

The SEM photomicrographs of cellulosic fibers showed an agglomerated aspect with fibers that were elongated and had thin edges. The cylindrical and well-defined shape of these fibers showed the presence of grooves along their surface, which gave them a rough aspect. Gaps were observed in the entire length of the walls of cellulosic fibers showing a range of heterogeneous sizes (Fig. 5a). GIC presented as a compact structure with a matrix involving heterogeneous particles (Fig. 5b) and the composite GICMF2 was also compact with random distribution of cellulosic fibers throughout the gelatinous matrix, in addition to load particles (Fig. 5c and d).

The fracture area of a representative specimen of GIC showed the presence of load particles heterogeneous in size (Fig. 6a). In the GICMF2 composite, in addition to GIC particles, cellulosic fibers were observed in the cementitious matrix (Fig. 6b and c). Fig. 6d and f suggested images of many particles adhered to the fiber surfaces. The swollen appearance of the cellulosic fibers within the ionomer cement mass may be observed in Fig. 6e.

The EDS spectrum of GICMF2 composite (Fig. 7) showed the presence of C and O, evident in high intensity peaks corresponding to the structure of the eucalyptus cellulosic fibers and Ionomer cementitious matrix; high intensity peaks of Ca, F, Al and Si, and low intensity peaks of Na and Ba corresponding to GIC load particles.

4. Discussion

The final properties of glass ionomer cement are influenced by the powder:liquid ratio and technique sensitivity [28–31]. A larger amount of load added to the cement and a higher powder:liquid ratio give the glass ionomer cement mechanical strength when compared with the traditional cement [20,32]. However, increased viscosity may hinder the handling of cement, in addition to affecting its bond to the tooth structure [27,28]. According to ISO 9917-1:2007 specifications, the changes in the ratio of powder–liquid of the glass ionomer cements may affect some of its properties [33]. A visible way of determining the working time of

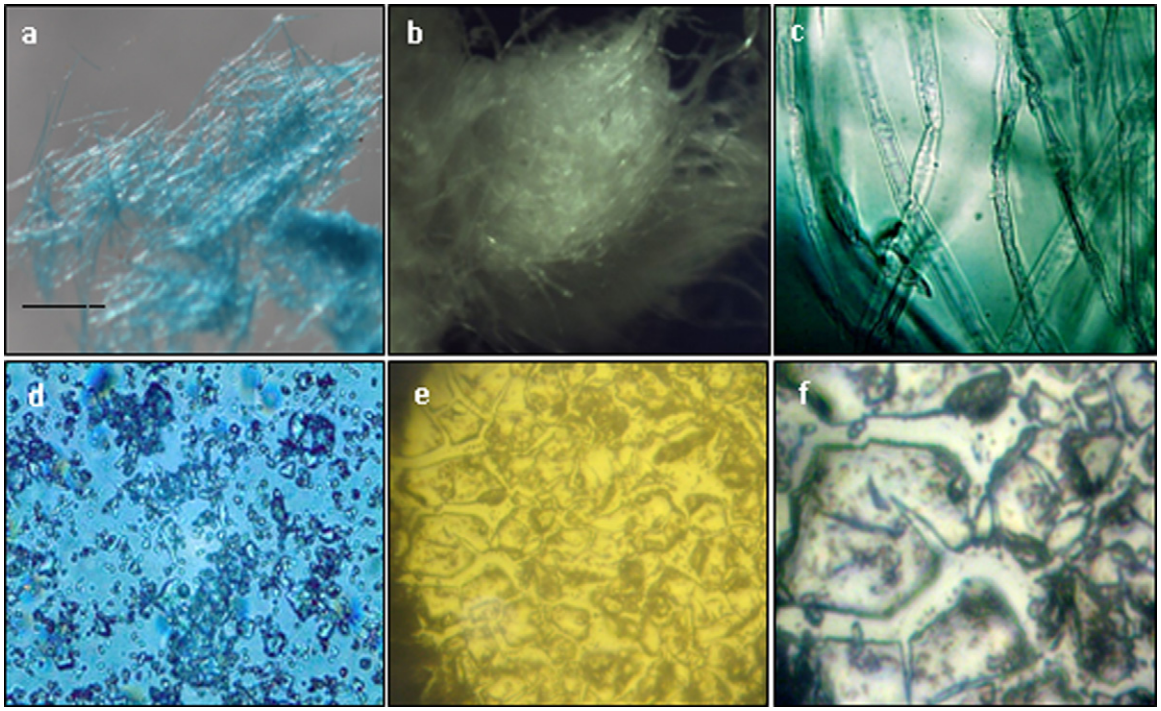

Fig. 4. Photos of eucalyptus cellulose fibers stained with Astra blue, before processing, at 45× magnification (a) and after processing, 45× magnification (b) in digital camera, cellulosic fibers after processing, 400× magnification (c) and GIC powder, 1000× magnification (d), stereoscope microscope, GIC powder, 3200× (e) and 5600× (f) magnification under inverted light microscope.

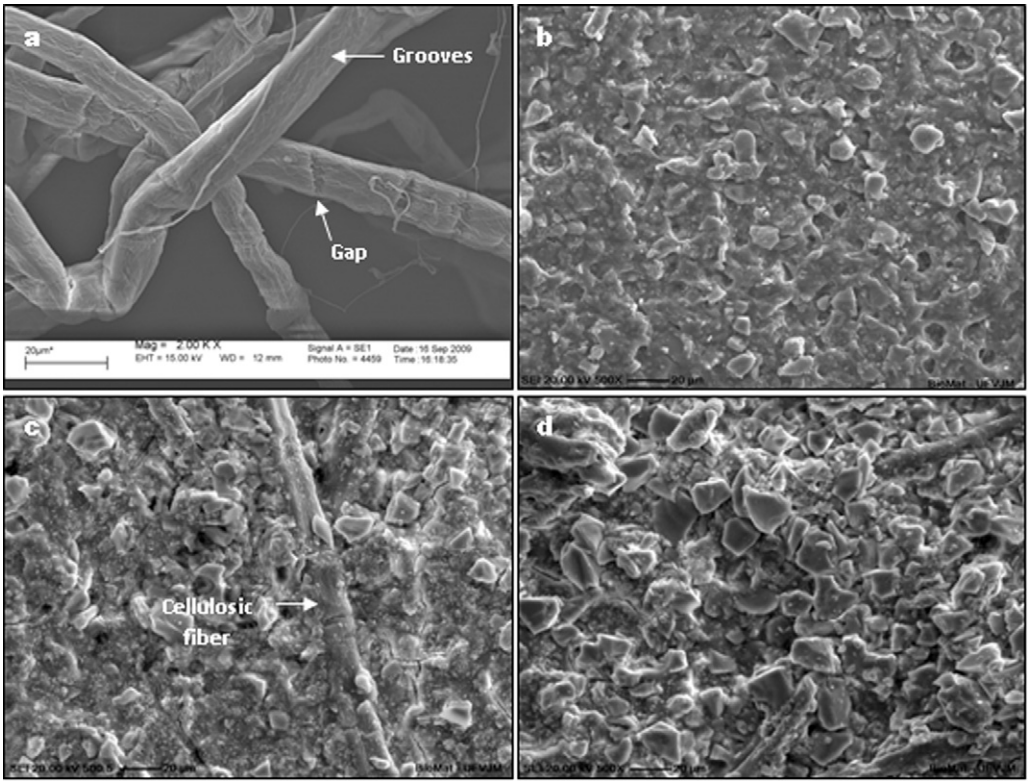

Fig. 5. SEM photomicrographs of the surface morphology of Eucalyptus cellulosic fibers, 2000× magnification (a), NMM/UFV, the surface of GIC, 500× (b); and the composite GICMF2, 500× (c and d), BioMat/UFVJM; 15 and 20 keV electron beam.

glass ionomer cement is by observing its shiny surface, indicating that non-reacted polyacids are present on this surface, and these are critical for cement bonding to the tooth [8,30,32,34]. The proportion of fibers added to GICMF1 and GICMF2 caused agglutination to occur easily in these cements, and provided working time similar to that of GIC, without changing their final setting time, thus respecting the characteristics of clinical relevance of this restorative material. The high concentration of fibers present in the matrix provided GICMF3 with viscosity that interfered negatively with its final properties.

In the first critical 24 h of setting time of glass ionomer cement there is greater susceptibility to loss and absorption of water. The high solubility and initial syneresis and imbibition are unfavorable properties, which may result in dimensional changes, loss of mechanical properties and formation of cracks [35,36]. Any contamination by water in this phase may cause dissolution of cations and anions in the matrix. Considering the need for water balance in glass ionomer cement in the initial phase of maturation, immediate protection of its surface against hygroscopic changes is recommended [31,37].

In this study, both the conventional glass ionomer cement used as control and those modified with fibers, with or without varnish protection, showed susceptibility to imbibition, especially in the first stage of maturity. Overall, the fiber has great capacity to retain water and because cellulose is a polymer with a high number of hydroxyl groups of a hydrophilic nature. The fact that the glass ionomer cement modified with fibers did not present a greater degree of hydration compared with conventional cement is explained by the formation of multiple hydrogen bonds between the hydroxyl groups of the different overlapping glucose chains that are important constituent of cellulose. Despite the fibrous structure of cellulose, water is capable of causing a limited increase in the volume of cellulosic fibers, making them virtually impervious to water and therefore insoluble, originating compact fibers [21,23,38,39]. Another factor to be discussed is the concentration of fibers added to the cement, which in this study did not contribute to increase in fluid retention.

According to Francisconi et al. [40], if glass ionomer cements are exposed to the air without any protection, their surfaces will suffer cracks and fractures as result of desiccation. After 7 days, the glass-ionomer cements modified with fibers and with varnish protection behaved in a similar manner to the conventional type with regard to syneresis. However, the composites GICMF3 and GICMF2 showed a higher degree of syneresis compared with the control group and the GICMF1 after longer exposure, 14 and 30 days. The higher concentration of fibers in cement seems to interfere with this process, with marked water loss and visibly characterizing the surface of materials with dryness and cracking.

Although glass ionomer cement modified with fibers presented significant solubility in comparison with the control group, it met the solubility requirements recommended by Specification No. 57 of the ADA [41], which states that the water solubility of a cement should not exceed 3% by weight of original mass and the sample must not present evidence of disintegration. The solubility of glass ionomer cement is related to the matrix gel [42], so it is assumed that the addition of fibers did not significantly affect the solubility of the materials. In addition, there was no disintegration on the cement surface under the conditions evaluated in this study.

Studies have shown increased strength in composites, after inclusion of cellulosic fibers in the material [23,45]. The compressive strengths of GICMF2 and GICMF1 composites were increased by modification of the physical structure with the incorporation of cellulosic fibers, and with regard to this property, the GICMF2 composite had the best behavior. The cellulosic fibers used in this study had a high incidence of pores from the gaps and lumens through which the sap of the eucalyptus moves [26]. According to the microstructural analysis, the mass of glass ionomer appears to be entrained within the fiber probably due to the porosity present

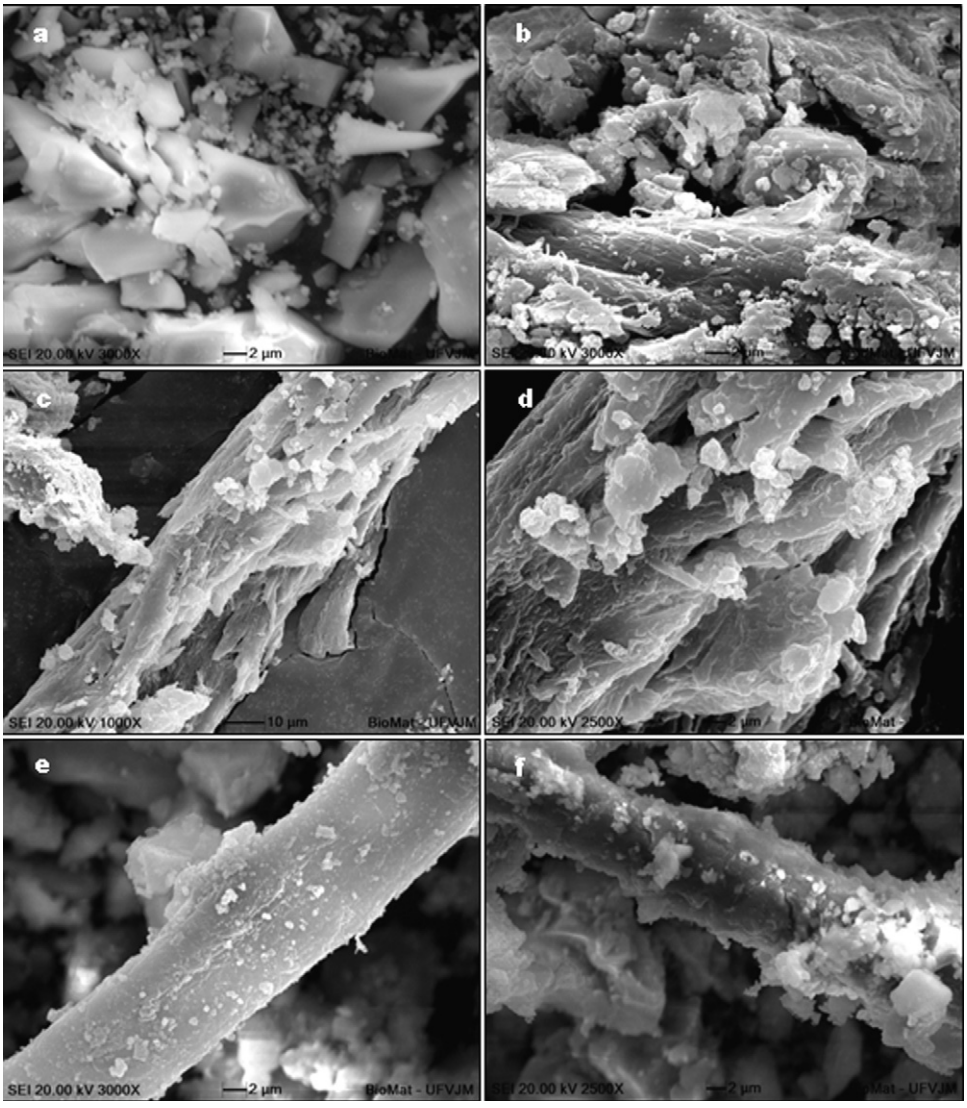

Fig. 6. SEM photomicrographs of GIC fracture areas, 3000× magnification (a), showing load particles, the GICMF2 composite: 3000× (b) and 1000× magnification (c), showing the cellulose fiber cement matrix 2500× (d and f), showing GIC load particles adhered to the fiber surface; 3000× (e), showing the cellulose fiber swollen within the cementitious mass; BioMat/UFVJM; 20 keV electron beam.

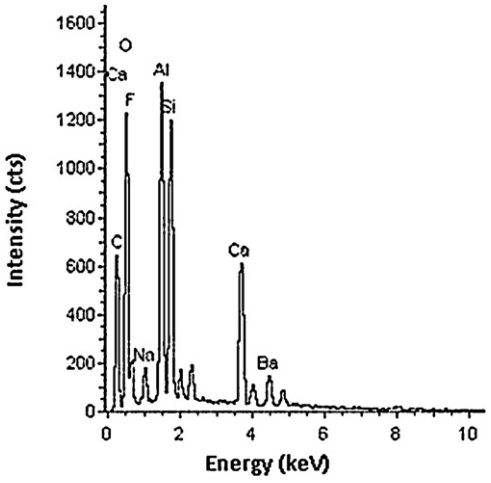

Fig. 7. EDS spectrum of the GICMF2 composite with high intensity peak of the main constituents of the cellulose chemical chain (C and O) and chemical constituents of the GIC calcium glass fluoraluminosilicate (SiO_2–Al_2O_3–CaF_2–Na_3AlF_6–$AlPO_4$).

in the fibers. Consequently it may have a filling of hollow fiber promoting increased interaction between the constituents of the composite. The state of high polymer concentration of crystalline cellulose present in the fibers may also have contributed to increasing the strength of glass ionomer cement modified with fibers [43]. When analyzing the results of this study, it can be observed that fiber concentration in the composite is a critical point in the final strength of the materials [44]. A lower concentration, such as that in GICMF1, entails structural weakness of the composite, since the small amount of fiber is unable to form a reinforcing network and thus being insufficient to absorb the tensile strength exerted. In higher concentrations, fibers can become entangled, as occurred in GICMF3, preventing the interposition of the cementitious matrix between them, leading to a potential point of fracture.

The diametral tensile strength remained unchanged in the composite, even in GIVMF2, which showed greater compressive strength. This is probably related to the fiber concentrations added to the composite. Glass ionomer cements are brittle, a property that limits their use as an option for restorative material in permanent teeth [30]. Thus, there is need for addition of cellulose

fibers in glass ionomer cement in an optimal concentration in order to increase the compressive strength and decrease friability, and retain a viscosity that facilitates its handling and does not interfere with its ability to wet the tooth structure for effective bonding.

The method of evaluating material wear by metallographic polishing is simple and accurate, and was supported by Harrington et al. [46]. Glass ionomer cement wear is related to the conventional characteristic of the matrix, which is formed by an acid-based crosslinking reaction [17]. In this study, fibers added to the ionomer cement matrices increased their resistance to wear. This fact suggests that an increase in the percentage of fibers in the matrix would provide a considerable increase in the rupture modulus of the original material, and confirms results of studies that observed the use of fibers as load material [21,23,47].

Pre-treatments for cellulose swelling by increasing their reactivity are common practices in the cellulose derivative industry. Swelling, especially the intra-molecular type, exposes the fibril surfaces and thus each fibril reacts along its length [23]. The addition of cellulose fibers to the composition of conventional restorative glass-ionomer cement increased its bond strength to dentin. Probably, the cellulose swelling with the polyacrylic acid solution increased the fibril reactivity with the cementitious matrix and tooth structure. There are many hydroxyl groups present in these fibers, due to their chemical structure [23,48]. This probably increased the chemical bonds between hydroxyl groups and calcium ions in dentin.

Characterization by SEM showed that GICMF2 was the most compact and homogeneous among the composites. After its setting time, there was a certain stability due to fiber aggregation in the manipulated cement paste. These morphological characteristics suggested the formation of a new composite with fiber/ionomer matrix/load particle interaction. The elementary chemical composition observed in the composite by means of the EDS spectra is consistent with the formation of calcium glass fluoraluminosilicate (SiO_2–Al_2O_3–CaF_2–Na_3AlF_6–$AlPO_4$) found in the literature, used in the manufacture of conventional restorative glass ionomer cement [30] and the chemical constitution of the cellulose chain $(C_6H_{10}O_5)_n$ [23,37].

The improvement in mechanical strength of glass ionomer cement modified with fibers will provide greater clinical longevity of restorations performed with this restorative cement. International agencies seek answers to the need for finding a method to preserve the teeth in individuals of all ages in underprivileged communities in developing countries, where resources are scarce. One technique indicated for this purpose is Atraumatic Restorative Treatment (ART), which is minimally invasive and seeks to preserve as much tooth structure as possible, using adhesive materials in restorations [49,50]. Glass-ionomer cements are the most common materials used in atraumatic restorations and provide benefits to dental health; however, one of their limitations is the low resistance in high-impact chewing regions [51]. Thus, the fiber-modified glass-ionomer cement could be used as definitive restorative material in the ART [52,53]. Therefore, it is expected to achieve simplification of the restorative procedure, plus social benefits, achieving increase in coverage to an unattended population, and reducing tooth losses. The GICMF2, which showed the best results in this study, could provide a feasible alternative restorative material for this purpose because, in addition to providing improved mechanical strength; its final cost should be much lower when compared with the materials with the same indication, found on the market.

The addition of cellulose fibers to glass ionomer cement modified the material structure so that it can be used in areas of great masticatory efforts, such as in restoration of posterior teeth, especially in the ART technique. Improvements in physical properties and more resistance testing should be performed in this cement. In the future, its clinical follow-up is planned to provide final short- and long-term proof of its performance. However, it is still necessary to perform compatibility and cellular cytotoxicity tests in vitro and in vivo evaluation in animal models before clinical use.

5. Conclusions

Based on the results obtained is possible to infer that:

- the glass ionomer cement modified with fibers, without or with protective varnish, showed no imbibition greater than that observed after 24 h, showing a water absorption capacity similar to that of GIC;
- within clinically acceptable limits, the ionomer cement modified with fibers had higher solubility in water than the GIC; none of the cements showed evident signs of disintegration;
- the addition of eucalyptus cellulosic fibers to the GIC, in the proportion used in GICMF2, provided higher compressive strength, resistance to abrasion and bond strength when compared with the GIC without fibers; however, the proportion of added fibers did not interfere with the composite diametral tensile;
- the morphological analysis and elementary chemistry of GICMF2 showed the formation of a new and stable composite with interaction between fibers/ionomer matrix/load particles.

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