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Influence of Multi-cycle Infiltration on Porosity and Optical Properties of Glassinfiltrated Alumina Biocomposites for Dental Restorations

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Abstract

Glass-infiltrated alumina biocomposites have been largely used in Dentistry due to their biocompatibility, high potential to mimic the natural dentition and good marginal adaptation. However, their optical and mechanical properties are jeopardized by the presence of residual porosity in the final structure. The objective of this study was to increase light transmittance of alumina-based biocomposites that are infiltrated with a lanthanum-containing aluminosilicate glass by means of increasing the number of infiltration cycles from one to three. The influence of the number of cycles on both residual porosity and optical properties was assessed. The infiltration cycles were carried out at 1200°C for 95 min. After each cycle the residual glass was removed and an additional glass layer was placed over the alumina preform. The SEM images showed that specimens infiltrated with three cycles had denser microstructure, with a residual porosity of 1.8 vol%, whereas specimens submitted to only one cycles showed residual porosity of 4.5 vol%. Transmittance increased 55% for specimens subjected to three infiltration cycles, while light scattering coefficient was reduced by 45% and the contrast ratio was reduced by 23%. These results indicate that the higher number of cycles resulted a more translucent material due to lower porosity and lower pore size (smaller than 3.2 µm). In conclusion, this work showed that multi-cycle infiltration is effective in reducing residual porosity and improving light transmittance of the biocomposite tested.

Keywords: Biocomposite, Glass, Alumina, Dental prostheses.

Introduction

Metal infrastructures veneered with porcelain are the most frequently used systems to prepare dental prostheses. Despite their high clinical success rate, reported to vary between 72 and 87% after 10 years, [1] inappropriate aesthetics due to opacity and allergic reactions to metals have led to the development of all-ceramic prostheses [2]. Good aesthetics for artificial teeth can be obtained using glass-infiltrated alumina biocomposites, which can be translucent enough to mimic the characteristics of the natural teeth, [3] yet keeping an excellent, accurate marginal and internal adaptation after multiple heat treatments [4]. The good marginal adaption of these glass-infiltrated alumina biocomposites is related to the low shrinkage of the final material, [5] which is below 0.5%, due to the fact that pre-sintering of the preform and infiltration of the glass are performed at relatively low temperatures [6].

The microstructure of glass-infiltrated alumina biocomposites is composed of a tridimensional network of two interlocked and continuous phases [7]. One of the phases is composed of alumina, which functions as a scaffold that is further infiltrated by a glass phase [2]. Although this interlocked microstructure provides good mechanical properties, it has problems related to the optical behavior of the restoration, as light is heavily scattered within the material, especially at interfaces due to a mismatch between the refractive indexes of both phases. Light scattering significantly decreases total light transmittance through the material [8]. In addition, residual porosity significantly reduces the mechanical properties of the biocomposite, [5] and is considered the most significant optical heterogeneity in its microstructure, causing significant light scattering and therefore decreasing light transmittance [8].

One possible solution for the problem of porosity in glassinfiltrated alumina biocomposites is repeating the infiltration cycle in order to improve glass penetration into the porous ceramic preform for porosity reduction. It seems that there is no work reported in literature showing the beneficial effects of carrying the infiltration cycle more than once on the optical properties of such biocomposites. Therefore, the aim of this work was to investigate the effectiveness of applying multi-cycle infiltration (i.e., repeating the procedure three times) on the porosity and the optical properties of a biocomposite for dental restorations.

Experimental

Glass preparation

The raw materials used for glass preparation were: quartz sand

(>99.5% SiO₂, Jundu, Brazil - L/N AQ-60-150); Al₂O₃ (99.9%, Showa Denko, Japan – L/N UA5105); boric acid (>99.5% H₃BO₃, PA ACS, Vetec, Brazil - L/N 1106670); TiO₂ (>99%, 1001, Kronos, USA - L/N 61040); and La₂O₃ (99.9%, PA, Vetec, Brazil - L/N 1306342). The powders were weighed using an analytical balance (AUY220, Shimadzu, Japan – S/N D312120166) aiming at the composition 25% SiO₂-20% B₂O₃-20% Al₂O₃ -25% La₂O₃-15% TiO₂ (mol%), considering the water loss from H₃BO₃ to B_2O_3 conversion during heating. The powder mixture (100 g) was first mixed using a mortar and a pestle, and then manually homogenized by rotating the powders inside a plastic bag inflated with air during 15 min. The mixture inside a 100 mL Pt-5%Au crucible was heated using an electric furnace (FE-1700, Fortelab, Brazil - S/N 004-2011) up to 1500 °C and holding at this temperature during 1 h for melting the powders. Then, the glass melt was removed from the furnace and immediately poured in distilled water (Figure 1) to obtain a frit (coarse glass powder), which was wet milled in a ball mill (MA500, Marconi, Brazil - S/N 101030104), using plastic jar, alumina balls and distilled water, in order to prepare a glass powder finer than 100 mesh (150 µm).



Figure 1: Image showing the quenching of melt glass in distilled water for the preparation of glass frit

Preform infiltration

In-Ceram Alumina blocks (CA-12, Vita Zahnfabrik, Germany - L/N 50840) were cut using a precision sectioning cutter (IsoMet 4000, Buehler, USA - S/N 719-1S4S-02053) with a diamond wafering blade, operating at 1000 rpm and 9 mm/min of automatic advance, and cooled with water flow, to prepare preforms with dimensions of 10.5 x 12.5 x 1.5 mm. Then, the preforms were washed in distilled water using an ultrasonic bath during 5 min (Eco-Sonics, Ultronique, Brazil - S/N UT13401020). For the glass infiltration process, the glass powder (~0.35 g) was mixed with a small amount (10 wt%) of distilled water to facilitate powder manipulation, and placed on the as cut preform surface. This set was placed on a platinum foil and then heat treated in an electric furnace (Kerampress, Kota, Brazil - S/N KP0811109) at 1200 °C during 95 min (maximum time allowed by the prosthetic furnace) under vacuum, with heating rate of 60 °C/min, corresponding to one cycle of infiltration. At the end

of the cycle, the furnace was switched off and the samples were removed from the furnace and bench cooled. The excess glass that remained on the preform was removed using emery papers. In order to evaluate the effect of number of infiltration cycles, two experimental groups were prepared: group 1 with samples prepared with one cycle; and group 2 with samples prepared with three cycles, repeating subsequently the steps described above for one cycle. Infiltration depth along the thickness was analyzed with an optical microscope in transmitted light imaging mode (SZ2 ILST, Olympus, Japan – S/N OL43751).

Both larger surfaces of the biocomposites were ground and polished to the thickness of 1.0 mm, using a semi-automatic grinder-polisher (EcoMet 250, Buehler, USA – S/N 719-E25G-D1584) with diamonds ranging from 45 to 1 μ m.

Characterization

For identification of crystalline phases, X-ray diffraction (XRD) analysis was performed using a diffractometer (D8 Focus, Bruker, USA - S/N 205085; Cu-Ka radiation) in the interval of 20 to 80° in 2 θ , using a step size of 0.05° and 2 s of counting per step. A scanning electron microscope (SEM, Quanta 600 FEG, FEI, USA - NC 943202034231) coupled with an energy dispersive electron spectrometer (EDS, Xflash Quanta 400, Bruker, USA) was used to analyze the microstructure of the biocomposites. For porosity determination of the alumina preform, first its apparent density was calculated by the geometric method: the ratio between the mass, measured in an analytical weighing scale (AUY220, Shimadzu, Japan – S/N D312120166), and the specimen volume, calculated from the dimensions measured with a caliper. Then, the ratio between apparent density and theoretical density of alumina (3.986 g/cm³), [8] resulted in the relative density of the preform. The volume fraction of pores (porosity) was calculated subtracting the relative density from 100% [8]. Pore fraction and size distribution of the biocomposites were determined analyzing SEM images of polished sections in the Image J software. Five images of different regions were used for each biocomposite.

For optical characterization, total transmittance and diffuse reflectance (using white and black backgrounds with glycerol as coupling liquid) of the polished biocomposites were measured using a spectrophotometer (CM-3700d, Konica Minolta, Japan – S/N 18916103) in the visible light region (360 to 740 nm) with 10 nm interval. To determine the contrast ratio (CR), an optical parameter that indicates the material's translucency, it was calculated the ratio of reflectance values measured with black and white backgrounds [9]. Using Kubelka-Munk (K-M) model, the absorption (K) and scattering (S) coefficients were determined [10,11].

Statistical analysis of the results was made using Student's *t*-test method at 95% confidence interval.

Results and Discussion

After milling, the glass powders had a homogeneous whitish appearance. The XRD pattern of the prepared glass did not show any sharp diffraction peak, characteristic of crystalline phase, but had only amorphous broad bands indicating a glassy structure (Figure 2). Before and after the glass infiltration process, the preform dimensions were measured with a caliper and no significant shrinkage was observed even after three infiltration





Figure 3: Transmitted light optical microscope images (light source at the back of specimens) of alumina preform before infiltration (left) and after one cycle infiltration (right), showing that the glass infiltrated through the entire thickness of preform. Note that only glass infiltrated region of the preform become light translucent (the upper and lower specimen surfaces are indicated with arrows). The dark grey region above the upper surface of preform in the top of the right image is the residual glass

cycles. The preforms were totally infiltrated by the glass melt along the thickness with just one cycle, as shown in Figure 3, but the residual porosity was different for the two groups tested.

Figure 4a shows a SEM image of the alumina preform (before infiltration), displaying alumina grains with broad particle size distribution and many pores, as expected. The total porosity of the preform was 25.5 vol%. SEM images of the polished biocomposite samples showed a microstructure with broad size

distribution of alumina particles surrounded by the infiltrated glass. A higher concentration of pores for specimens infiltrated with one cycle can be seen in Figure 4b, indicating that only one cycle of 95 min was not enough for the glass to penetrate and fulfill all the pores inside the preform. After three infiltration cycles a reduced fraction of residual pores was observed (Figure 4c). EDS analysis showed the presence of Al an O in the dark gray regions in micrographs of Figure 4b and 4c, which correspond to alumina particles, and the presence of Si, Al, La, Ti and O elements in the light gray regions, corresponding to the glass phase. The absence of boron in the analysis of glass matrix was due to the insensibility of this technique to this element.

For specimens infiltrated with one cycle, residual porosity reached 4.5 vol%, while after three cycles, the porosity level was 1.8 vol% (Figure 5), representing a reduction of 60% in total pore fraction. Pore size distribution was analyzed and the results are



resent one standard deviation. Values followed by different letters are statistically different



Figure 6: Pore fraction as a function of pore size range (in μ m)



Figure 4: SEM images of preform surface (a), and polished sections of biocomposites infiltrated with one cycle (b) and three cycles (c). The arrows indicate the pores. In (b,c), light grey regions are the infiltrated glass, and dark gray regions are the alumina particles

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shown in Figure 6. After three infiltration cycles, all pore sizes were reduced, however the reduction of pores with less than 3.2 μ m was more intense.

Capillary pressure is the driving force responsible for glass infiltration into the pores of the preforms, and it is inversely proportional to the capillary (pore) radius [12,13]. As it can be seen in the SEM image (Figure 4), the regions in the preform filled by the glass were thinner than those that were not filled (residual non-infiltrated pores), especially in Figure 4b (with one cycle). After one cycle, the fraction of smaller pores was still high when compared to the larger ones (Figure 6), but after three cycles, the fraction of smaller pores was significantly reduced. These results indicated that smaller pores were filled by the glass before the larger ones, probably due to the higher capillary pressure in smaller pores. Similar result was also observed in the literature [14], although some works reported a contrary effect [15].

Light transmittance results for samples prepared with one infiltration cycle reached 22% whereas samples prepared with three infiltration cycles reached almost 34% of transmittance (Figure 7). This difference corresponds to an increase of 55% in the wavelength region between 450 and 750 nm. Light scattering (S) and absorption (K) coefficients of the biocomposites were calculated from the reflectance values measured on white and black backgrounds. The scattering coefficient was higher for the biocomposites prepared with one infiltration cycle and had a significant reduction of about 45% for the sample infiltrated with three cycles (Figure 8a). The light absorption coefficient was not significantly affected by the number of infiltration cycles (Figure 8b). The higher transmittance measured for the samples infiltrated in three cycles in all visible wavelength region (360 to 740 nm) was mostly due to the lower scattering coefficient in this region. Therefore, as it can also be observed in Figure 7, transmittance was strongly reduced for wavelengths below 450



nm in both the cases, due to the intense increase in the light absorption coefficient in this wavelength range.

A light beam is usually scattered when it crosses an interface between two phases with different refractive indexes due to refraction, leading to diffuse transmittance through a multiphase material. Considering the residual non-infiltrated pores as a (gas) phase, they can reduce the translucency of the material due to strong light scattering at the interface between solid and gas (pore).

Light scattering in optically heterogeneous systems, which can be assumed as a transparent medium with small particles [16], can be modeled by Mie solution to Maxwell's equations for particles of arbitrary size and refractive index relative to the medium [17]. This theory shows that scattering is a strong function of particle size and maximum scattering occurs when the particle size is close to the wavelength of the incident radiation [18]. The higher transmittance seen in specimens infiltrated with three cycles can be attributed to the reduction in residual porosity and also in pore fraction with diameter smaller than 3.2 μ m. The lower light scattering coefficient estimated for biocomposites infiltrated with three significantly reduce the translucency of the material.

Furthermore, it is important to consider the scattering that occurs at the interface between alumina and glass, due the difference between the refractive indexes of these phases. The scattering phenomenon is expected to be less intense when these indexes are similar. The refractive index of the glass investigated in this work, measured in a refractometer (2010/M Prism Coupler, Metricon), was 1.7411, which is 1.1% lower than that obtained for alumina (1.76). The glass composition used in this study was designed by a systematic approach developed by this research group, [19] aiming to minimize the difference between the refraction indexes of glass and ceramic phases in the biocomposite to achieve high transmittance.

The contrast ratio (CR), obtained from the reflectance values, for the specimen prepared with one cycle was 0.798, whereas for the sample prepared with three cycles the CR was 0.615 (Figure 9). Lower CR values correspond to higher translucency of the material, therefore, this result also confirmed the better optical properties of the specimens prepared with three infiltration cycles compared to one cycle, reaching a reduction of 23% in the CR value.

Infiltration time is an important parameter to be considered in order to obtain a homogeneous microstructure which provides





better optical properties due to the elimination of regions with different refractive indexes. Although the glass had already infiltrated the whole thickness of the preform with one cycle of 95 min, this infiltration time was not enough for the glass to infiltrate in all pores inside the preform, once the capillary pressure is reduced in larger pores [12,13]. The fact that the infiltration distance has a parabolic dependence on time suggests that, if the sample thickness is doubled, the time for total infiltration would be four times higher, and it would have to be carefully estimated in order to result in a good prosthesis.

Conclusions

The results of this work confirmed the effectiveness of applying multi-cycle infiltration (repeated three times) to reduce the porosity and to increase the optical properties of a biocomposite for dental restoration. Infiltration was complete through the thickness with just one heat treatment of 95 min, however, there were still a lot of residual pores in the structure. Residual porosity was reduced in 60 vol% after three cycles, reducing mostly pores with less than 3.2 µm of diameter, which indicates that smaller pores were filled before larger ones, probably due to the higher capillary pressure in smaller pores. The transmittance increased 55% for the samples processed with three infiltration cycles, the scattering coefficient reduced about 45% and the contrast ratio value reduced 23%, resulting in a more translucent material, due to the reduction in pores with sizes within the visible spectrum wavelength range. Infiltration time significantly affects the optical properties, due to the elimination of the regions with different refractive indexes, and it needs to be carefully estimated in order to obtain acceptable dental prosthesis.

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