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Systematic approach to preparing ceramic–glass composites with high translucency for dental restorations

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ABSTRACT

Objective. Ceramic composites are promising materials for dental restorations. However, it is difficult to prepare highly translucent composites due to the light scattering that occurs in multiphase ceramics. The objective of this work was to verify the effectiveness of a systematic approach in designing specific glass compositions with target properties in order to prepare glass infiltrated ceramic composites with high translucency.

Methods. First it was necessary to calculate from literature data the viscosity of glass at the infiltration temperature using the SciGlass software. Then, a glass composition was designed for targeted viscosity and refractive index. The glass of the system SiO_2–B_2O_3–Al_2O_3–La_2O_3–TiO_2 prepared by melting the oxide raw materials was spontaneously infiltrated into porous alumina preforms at 1200 °C. The optical properties were evaluated using a refractometer and a spectrophotometer. The absorption and scattering coefficients were calculated using the Kubelka–Munk model.

Results. The light transmittance of prepared composite was significantly higher than a commercial ceramic–glass composite, due to the matching of glass and preform refractive indexes which decreased the scattering, and also to the decrease in absorption coefficient.

Significance. The proposed systematic approach was efficient for development of glass infiltrated ceramic composites with high translucency, which benefits include the better aesthetic performance of the final prosthesis.

1. Introduction

The most widely used system to produce fixed partial dentures (FPDs) is the metal–ceramic, which is constituted of a metallic framework veneered with a dental porcelain. High clinical success rates for this system have been reported, varying between 72% and 87% after 10 years [1]. However, the opacity of the metal jeopardizes the final aesthetic outcome of the prosthesis, as it is difficult to mimic the translucent

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dental tissues [2]. Such aesthetic drawback pushed the development of all-ceramic systems, in which the framework can be built with a glass–ceramic, a glass–infiltrated ceramic composite or a polycrystalline ceramic [3]. In comparison to metals, these ceramic materials have excellent biocompatibility, high color stability, and low thermal conductivity [4]. However, it was observed that for most commercial dental ceramics, the higher the mechanical strength, the lower the translucency [5]. Therefore, it is difficult to obtain ceramic frameworks with high mechanical properties and acceptable aesthetic qualities at the same time.

Glass-infiltrated ceramic composites are promising materials that have the potential to fulfill the requirements of having both high toughness and high translucency. Nevertheless, unfortunately the commercial glass–infiltrated composites currently available in the dental market do not fulfill these requirements [5]. These materials still have low translucency due to an important mismatch of the refractive indexes of the different microstructural phases that results in light scattering at the interfaces, decreasing light transmittance through the body [6]. Indeed, ordinary silicate glasses usually have low refractive indexes ($n = \sim 1.5$) compared to most reinforcing oxide phases, like alumina, spinel ($MgAl_2O_4$) and ceria-stabilized zirconia ($n = 1.7$–2.2). The addition of titanium oxide and lead oxide can significantly increase the refractive index of silicate glass and help solving the light scattering problem in these composites [7], but because of its toxicity, it is not safe to use lead oxide in biomaterials. Some aluminosilicate glasses containing rare earth oxides have relatively high refractive indexes ($n = 1.7$–1.8), especially after the addition of lanthanum and yttrium oxides [8–10]. This explains why glasses that are infiltrated into alumina preforms usually contain lanthanum oxide [11]. Another important glass property is the viscosity, since it affects the temperature at which the infiltration process is carried out. However, the literature lacks information about the viscosity values of glasses in the range of the infiltration temperature.

The aim of this study was to verify the effectiveness of a systematic approach using a software that takes into account the glass properties to predict the processing parameters related to glass infiltration. This work also investigated the capability of this systematic approach in designing specific glass compositions with target properties in order to prepare glass infiltrated ceramic composites with high translucency.

2. Materials and methods

In this work, alumina (Al_2O_3) was chosen as the ceramic phase for the composite preform because of its good mechanical properties (421 GPa of Young's modulus, 4.0 MPa m$^{1/2}$ of fracture toughness, 397 MPa of four-point flexural strength, and 19.8 GPa of Vickers hardness), higher than spinel ($MgAl_2O_4$) ceramic [12,13]. Although the preform of zirconia–toughened alumina results in a stronger composite [14], it is difficult to match the refractive index of the infiltrating glass to both ceramic phases, since they have significantly different refractive indexes (1.76 and 2.19 for alumina and zirconia, respectively [8]).

2.1. *Simulation of glass properties and compositional design*

The software SciGlass 7.7 (Glass Property Information System, Lhasa) was used to calculate the viscosity and refractive index of the glass from its chemical composition. In order to design the glass compositions, it was necessary to know the viscosity of the glass at the infiltration temperature, in order to restrict the maximum temperature to 1200 °C, which is the capacity of commercial furnaces for this process. Therefore, it was necessary to calculate this viscosity based on the data provided by the literature. Fig. 1 shows the calculated viscosities from the chemistry of some glasses as a function of the temperature used for the infiltration process reported in Refs. [15–17]. It could be observed that the viscosities of the glasses at the infiltration temperature ranged from around 10^2 to 10^3 dPa s. Therefore, in this work it was established that the glass should have viscosity lower than 10^2 dPa s at 1200 °C for the design of glass composition.

The basic glass composition chosen was 45% SiO_2, 25% Al_2O_3, 15% La_2O_3, and 15% TiO_2 (mol%), since a similar composition was reported [9] to result in glass with refractive index close to that of alumina. The simulation of this composition with software SciGlass 7.7 indicated viscosity of 10^2 dPa s at 1422 °C for this glass, but this temperature was too high for the scope of this work. In this way, the addition of alkaline oxides (Li_2O, Na_2O, K_2O, Rb_2O, and Cs_2O) was simulated in order to replace silica in this glass, however instead of decreasing the temperature, this change resulted in an increase in the temperature at the viscosity of 10^2 dPa s (Fig. 2a). The simulations showed temperature dependence of viscosity that was similar for all alkaline oxides up to the addition of 25 mol% (Fig. 2b). The addition of most of the alkaline-earth oxides (MgO, CaO, SrO, and BaO) to the basic glass composition also increased the temperature at the viscosity of 10^2 dPa s (Fig. 2c), except for the addition of beryllium oxide which lowered this temperature to 1319 °C for 25 mol% addition (Fig. 2d), but not sufficiently to the target temperature (<1200 °C).

The simulations, however, indicated that the addition of boron oxide could significantly lower the temperature at

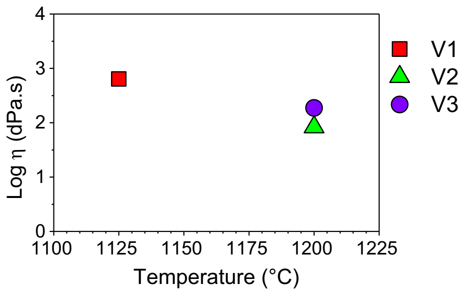

Fig. 1 – Viscosity calculated from the chemical composition of the glasses as a function of the temperature reported in the literature for the infiltration process into alumina preform (mol%): V1 – $41SiO_2$–$18B_2O_3$–$17Al_2O_3$–$11La_2O_3$–$5TiO_2$–$8CaO$ [15]; V2 – $33SiO_2$–$23B_2O_3$–$18Al_2O_3$–$13La_2O_3$–$7TiO_2$–$4CaO$–$2CeO_2$ [16]; V3 – $32SiO_2$–$23B_2O_3$–$17Al_2O_3$–$13La_2O_3$–$7TiO_2$–$5CaO$–$3CeO_2$ [17].