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Characterization of rare earth oxide-rich glass applied to the glass-infiltration of a ceramic system

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Abstract

The viability of a silica glass containing rare earth oxides as infiltration agents in different ceramic substrates was investigated. $ZrO_2(Y_2O_3)$–Al_2O_3 and Al_2O_3–$ZrO_2(Y_2O_3)$ composite ceramics were sintered at 1530 °C/2 h and characterized by X-ray diffraction (XRD), dilatometry and atomic force microscopy (AFM). The wetting behavior of the substrates by rare earth glass was studied by the sessile drop method at temperatures of up to 1285 °C in an argon atmosphere. Both composites presented high relative density (close to 98%) with α-Al_2O_3 and tetragonal ZrO_2 as crystalline phases. The wetting angle of the two substrates decreased in response to increasing temperature, reaching a final contact angle of 12.7° on the $ZrO_2(Y_2O_3)$:Al_2O_3 substrate at 1285 °C and of 13.6° on the Al_2O_3:$ZrO_2(Y_2O_3)$ substrate at 1275 °C, indicating good wettability in both cases. Results of fracture toughness show KIC of 4.3 MPa m1/2 and 5.4 MPa m1/2 for $ZrO_2(Y_2O_3)$:Al_2O_3 and Al_2O_3:$ZrO_2(Y_2O_3)$ respectively. The theoretical residual stress in the two infiltrated composites were calculated based on the coefficient of thermal expansion of the substrates and glass. The $ZrO_2(Y_2O_3)$:$Al2O_3$ and Al_2O_3:ZrO2(Y_2O_3) composites showed calculated residual stresses of 36.5 MPa (tensile) and 252 MPa (compression), respectively, indicating that compressive residual stress contributes to increase the toughness of the glass-infiltrated composites.

Keywords: Wettability; Zirconia-alumina composites; Residual stress; Interface; Glass characterization

1. Introduction

A trend in dentistry is to substitute metal-based prostheses for ceramic materials, mainly due to esthetic reasons. Alumina-based ceramics, Al_2O_3, have high hardness and wear resistance, while yttrium stabilized zirconia-based ceramics, $ZrO_2(Y_2O_3)$, have high fracture toughness and strength and are esthetically more attractive [1]. The high fracture toughness of $ZrO_2(Y_2O_3)$ ceramics is due to the tetragonal-to-monoclinic phase transformation, accompanied by a 3 to 6 vol% expansion in grains. The stress-induced tetragonal-to-monoclinic ZrO_2 transformation ahead of a crack tip generates compressive stresses in the ceramic matrix, hindering crack propagation and thus resulting in high fracture toughness [2,3]. High toughness is a very important factor in dental materials due to the induced and alternating stresses to which they are subjected [1]. During mastication the loads are usually in the order of 200 N, but may reach up to 1200 N [4].

A method widely used to produce dental prosthesis is by CAD/CAM machining of pre-sintered ceramic blocks with subsequent infiltration of a glass into a porous substrate and final sintering to densification. In this case, it is important that the coefficient of thermal expansion (CTE) of the ceramic

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substrate and the glass present only slight differences in order to minimize thermal mismatch, which could lead to undesirable internal stress and cause cracking of the component during processing. Another important parameter is the contact angle between glass and substrate, which indicates the wettability of the glass in the ceramic and the possibility of full densification after infiltration.

The mechanical stability of dental prostheses is a critical factor, because the durability of the junction must be ensured. Zirconia-based ceramics may undergo tetragonal-to-monoclinic ZrO_2 phase transformation in aqueous or salty environments. An alternative to monoclinic ZrO_2-based ceramics, which does not involve tetragonal-to-monoclinic ZrO_2 phase transformation, are composite materials based on ZrO_2–Al_2O_3 mixtures [2,3] or ZrO_2-glass systems [5].

In the production of composites with phases exhibiting distinct characteristics, as in the case of ZrO_2–Al_2O_3 infiltrated by a glass, it is necessary to study the physical and chemical interactions between the constituents. For effective infiltration, the glass must wet the surface of the substrate. This behavior is commonly described by the wetting or contact angle, θ [6–9]. This contact angle is established as a result of the interfacial energies between the liquid and solid phases [10–13]. Wettability is described by Young′s Eq. (1).

$$-\Delta G = \gamma^{lv}(1 + \cos\,\theta) \tag{1}$$

where ΔG represents the Gibbs free energy and γ^{lv} represents the surface energies of liquid–vapor.

If θ equals zero, wetting is maximum and spreading of the liquid occurs; when θ equals 90°, partial wetting occurs, and when θ equals 180°, wetting does not occur. Therefore, a minimum contact angle is necessary for wetting to occur.

The most common method for determining wettability is the sessile drop method [6,14–18]. This method consists of heating a fusible material on a solid substrate and measuring the contact angle of the liquid drop on the substrate as a function of temperature and/or time. The variation in the height and diameter of the liquid drop is recorded continuously with a camera.

Based on the thermal expansion coefficients of the matrix and glass phase, residual stresses are generated between the substrate and the glass [19], as illustrated in Fig. 1

- $\alpha_{glass} > \alpha_{substrate}$: shrinkage of the matrix (substrate) is higher than that of the glass. The matrix/glass interface is thus stressed, leading to radial microcracks in the matrix around the glass.
- $\alpha_{glass} = \alpha_{substrate}$: no stresses appear at the interface, since the matrix and glass shrink equally.
- $\alpha_{glass} < \alpha_{substrate}$: shrinkage of the matrix (during cooling after sintering) is lower than that of the glass. To improve the fracture toughness, the intensity of the tensile stress must be limited to prevent partial or total interfacial debonding.

The average thermal residual stress generated during the cooling of sintered samples is calculated based on the assumption of a homogeneous distribution of secondary phase in the ceramic matrix obtained during infiltration, and is directly related to the difference in the coefficients of thermal expansion of the substrate and the glassy intergranular phase [20–22]. The average residual stress in both phases can be calculated as a function of the volume fraction of secondary phase, based on the approach proposed by Shi et al. [21], using Eqs. (2) and (3).

$$\sigma_g = E_g(\langle\alpha\rangle - \alpha_g)\Delta T \tag{2}$$

$$\sigma_m = E_m(\langle\alpha\rangle - \alpha_m)\Delta T \tag{3}$$

Here, σ_g and σ_m are residual stresses in the system (glass and substrate matrix, respectively). E_m and E_g indicate the Young modulus of the matrix and glass, respectively, and α, α_m and α_g indicate the average coefficient of thermal expansion (CTE) of the composite, matrix and glassy phase, respectively. The average coefficient of thermal expansion for each composition can be calculated using Eq. (4)

$$\langle\alpha\rangle = \frac{\alpha_g C_g E_g + \alpha_m C_m E_m}{C_g E_g + C_m E_m} \tag{4}$$

where C_g and C_m are, respectively, the fraction of glass and matrix. Based on the above calculation one finds that, on average, when $\alpha_m > \alpha_g$ and $\sigma_g < 0$, the grain boundary will be in compression and the matrix will be in tension [21,22]. Residual stress in multiphase composites develops due to the mismatch of the E-modulus and the coefficient of thermal expansion (CTE) in the constituent phases. Due to the lower CTE of the glass α_b than that of the matrix α_m, residual tensile stresses develop in the ZrO_2–Al_2O_3 matrix during cooling.

The purpose of this work was to investigate the compatibility of the rare earth oxide-rich glass with $ZrO_2(Y_2O_3)$–Al_2O_3 and Al_2O_3–$ZrO_2(Y_2O_3)$ ceramic composites based on an analysis of wettability and residual stress. The analysis

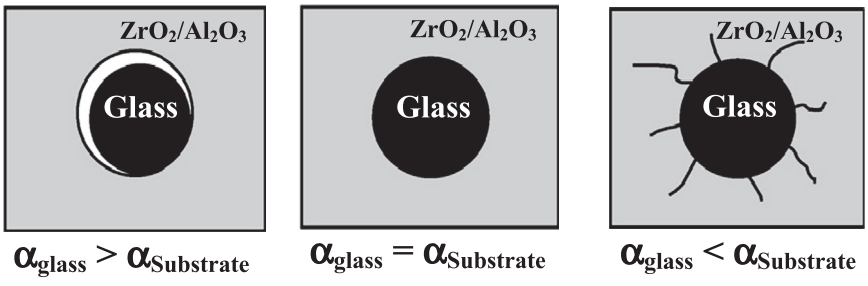

Fig. 1. Internal stresses and associated damage mechanisms involved during cooling by thermal expansion mismatch between spherical glasses embedded in a substrate.