Aging of 3Y-TZP dental zirconia and yttrium depletion

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ABSTRACT

Objective. Yttrium-stabilized zirconia is susceptible to low temperature degradation after interaction with water. Various mechanisms by which water molecules destabilize the tetragonal phase have been proposed, while the concept of yttrium depletion by the incorporation of hydroxyl ions in the crystalline structure either through the formation of Y—OH/Zr—OH bonds or small α-Y(OH)$_3$ crystallites, is prevailing. The present study was performed to investigate the surface alterations on a 3Y-TZP dental ceramic during the process of in-vitro aging and to further explore the yttrium depletion mechanism that occurs upon interaction with water.

Methods. Surface structural changes of zirconia specimens where investigated before and after in-vitro aging with X-ray diffraction analysis, Fourier-transformed infrared spectroscopy, X-ray photoelectron spectroscopy, fluorescence microscopy and scanning electron microscopy.

Results. High luminescence generated from the non-aged specimen was explained by the high amount of oxygen vacancies. The phase transformation from the t-ZrO$_2$ to the m-ZrO$_2$ phase after aging was accompanied by a significant loss of yttrium, a clear decrease of oxygen vacancies and a profound decrease of luminescence. Surface oxygen vacancies either migrated into the inner of the specimens or/and/engaged oxygen from the ZrO$_2$ and formed the metallic phase of Y$_2$O$_3$ on the surface after aging.

Significance. An "ideal" amount of oxygen vacancies that could stabilize the tetragonal phase in Y-TZP zirconia ceramics, without compromising esthetics and LTD resistance, is still a matter of further research and different susceptibilities to LTD among various dental zirconia ceramics are based on the amount of oxygen vacancies that can be annihilated by water molecules.

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1. Introduction

To date, stabilized zirconia is a well-established biomaterial used in dentistry. Zirconia used in dental applications is usually stabilized with 3 mol% Y_2O_3 (3Y-TZP) [1–3]. The use of Y_2O_3 was first suggested with the addition of 2–3mol% Y_2O_3 resulting in materials with only the desired tetragonal crystallographic phase ($t-ZrO_2$), called tetragonal zirconia polycrystals (TZP) [4]. The 3Y-TZP zirconia presents advanced mechanical properties as well as chemical, thermal and dimensional stability, compared to other ceramic systems.

Under stress, a spontaneous transformation takes place, from the high-energy state of the $t-ZrO_2$, which is present at temperatures between 1170 °C and 2370 °C, to the lowest energy state of the monoclinic ($m-ZrO_2$) phase, present at room temperature. This $t \rightarrow m$ transformation, known as a "transformation toughening mechanism" [5], is followed by a 3–4% volume expansion as a result of the larger volume occupied by the monoclinic phase, compared to the tetragonal. This expansion is considered to protect the zirconia ceramics from failure by restricting the growth and propagation of any surface microcracks inside the bulk of the material [5,6].

The crucial complication of Y-TZP ceramics occurs when the transformation proceeds in the presence of moisture and is known as aging or low-thermal degradation, (LTD) first reported by Kobayashi et al. [7]. The transformation is initiated at isolated grains on the surface by a stress corrosion type mechanism and when the entire surface is degraded, the degradation develops into the bulk of the material by micro and macro cracking [8], reducing its mechanical properties [9].

Up to date, investigations have attempted to explain this phenomenon and the following mechanisms have been proposed. Chevalier and his co-authors in 2009 suggested that O^{2-} from the segregation of water plays the main role for the filling of oxygen vacancies present in the structure and leads to the destabilization of the tetragonal zirconia [10]. Sato and Shimada [11] and Yoshimura et al. [12] also proposed that water reacts with the Zr—O bond and due to the movement of —OH at the surface and in the lattice, Y—OH and Zr—OH bonds are formed that trigger the $t \rightarrow m$ transformation. Lange et al. [13] postulated that water reacts with Y_2O_3 to form clusters of small (20–50 nm) α-$Y(OH)_3$ crystallites and induce depletion of the stabilizer. It was assumed that this reaction reproduces a monoclinic core (depleted of Y_2O_3) on the surface of an exposed tetragonal grain. Finally, they concluded that the above mechanism is promoted firstly because $Y(OH)_3$ is more resistant than Y_2O_3 in a hydrothermal environment and secondly because yttrium is less active in the degraded zirconia structure [13].

The understanding of the aging process and its influence is crucial for the future of zirconia as an acceptable dental material. Therefore, the present was performed to investigate the surface alterations on a 3Y-TZP dental ceramic, during the process of in-vitro aging, and to further explore the yttrium depletion mechanism that occurs upon interaction with water. It was hypothesized that the in-vitro aging of of 3Y-TZP zirconia specimens for 5 h and 10 h results in significant yttrium depletion from the superficial layers of the material's structure that facilitates the $t \rightarrow m$ transformation.

2. Materials and methods

2.1. Manufacturing of the specimens

A total of 15 specimens were utilized derived from a 3Y-TZP zirconia ceramic (IPS e-max ZirCAD, Ivoclar-Vivadent). Specimens with final dimensions (after sintering) 8×2 mm were prepared from zirconia blocks via CAD/CAM technology. Sintering was performed using the Vita zircomat T furnace, according to the manufacturer's instructions.

2.2. Aging of zirconia specimens

For the aging of the zirconia specimens, the model proposed by [10] was utilized, with aging time set at 5 and 10 h in an autoclave (KavoKlave 2100, KavoDental, Biberach/Riss, Germany). After in vitro aging, the specimens were dried with hot air and preserved until use in a desiccator, at room temperature (RT).

2.3. X-ray diffraction analysis (XRD)

The surface of three specimens from each group (untreated, after 5 h of aging and after 10 h of aging) was evaluated by XRD. XRD analysis was carried out using a diffractometer with Ni-filtered CuKα radiation (PW1710, Philips, Eindhoven and Almelo, The Netherlands). Diffraction patterns were obtained from 0° to 75° at a scan speed of 0.008°/min. The monoclinic phase fraction Xm was calculated according to the Garvie and Nicholson equation [14]:

$$X_M = \frac{I_{M(111)} + I_{M(11\bar{1})}}{I_{M(111)} + I_{M(11\bar{1})} + I_{T(111)}} \tag{1}$$

The yttria content of the $t-ZrO_2$ was calculated using the formula [15,16]:

$$YO_{1.5} (mol\%) = \frac{1.0225 - \frac{c}{a\sqrt{2}}}{0.0016} \tag{2}$$

2.4. Fourier transform infrared spectroscopy (FTIR)

The surface of the specimens analyzed with XRD was additionally evaluated by FTIR. The FTIR reflectance spectra of the specimens were obtained in the spectral area of MIR and FIR ($4000\,cm^{-1}$–$400\,cm^{-1}$) with a resolution of $2\,cm^{-1}$ utilising a Bruker FTIR extended spectrometer (model IFS113v).

2.5. X-ray photoelectron spectroscopy (XPS)

The XPS measurements were performed for each aging time (0h–5h–10h) on one arbitrarily selected specimen analyzed with XRD and FTIR. Measurements were carried out utilising a Kratos Axis Ultra DLD instrument, which uses a monochromatic Al–Ka X-ray beam and a hemispherical sector analyzer (HSA). The pass energy used to acquire the core level spectra was 20 eV. The spectra were corrected in terms of charging based on the peak of the C-1s (derived from the adventitious carbon on the surface) at 284.6 eV. The measured elements with their respective core levels were: C-1s, O-1s, Zr-3d and