Acta Materialia 144 (2018) 245–256

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Full length article

Towards the prediction of hydrothermal ageing of 3Y-TZP bioceramics from processing parameters

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ARTICLE INFO

Article history:
Received 19 June 2017
Received in revised form
25 October 2017
Accepted 27 October 2017
Available online 27 October 2017

Keywords:
Zirconia
Hydrothermal ageing

ABSTRACT

Hydrothermal ageing of yttria-stabilized tetragonal zirconia ceramics can have a strong influence on the lifetime of zirconia devices. Ageing kinetics are often described by the Mehl–Avrami–Johnson equation, most often used as a phenomenological description. This work seeks to relate the parameters of MAJ equations (V_{max}, n, b_0 and Q) to microstructural characteristics of the zirconia material: grain sizes, Y_2O_3 partitioning, monoclinic, tetragonal and cubic phases ratio. Samples with identical nominal composition of 3Y-TZP were prepared with grain sizes ranging from 190 nm to 773 nm. From their microstructural parameters, a relationship between microstructural parameters and sintering cycles was first proposed, followed by a relationship between ageing parameters and microstructural parameters. These results provide a convenient framework to better develop the sintering cycle of zirconia biomaterial in order to maximize their resistance to hydrothermal ageing.

1. Introduction

Zirconia-based materials, especially yttria-stabilized tetragonal zirconia polycrystals (Y-TZP), exhibit the best mechanical properties of oxide ceramics: this is the consequence of phase transformation toughening, which increases its crack propagation resistance [1–3]. Furthermore, they are high-temperature resistant and offer low thermal conductivity, good ionic conductivity, good biocompatibility and superior aesthetic appearance [2]. Therefore Y-TZP ceramics are very attractive for a wide range of biomaterial applications such as fixed-partial denture in restorative dentistry or femoral heads in orthopedics [4,5]. Large efforts were already devoted to the study of mechanical properties of zirconia-based ceramics, but also of their sensitivity to low temperature degradation (LTD). Low-Temperature Degradation of zirconia (also called hydrothermal ageing) can be summarized as follows: the presence of water in the environment of zirconia pieces triggers the tetragonal to monoclinic (t–m) transformation of some grains on the surface, which potentially leads to roughening and micro-cracking because of the high volume increase (~5%) associated to the t-m transformation. Hydrothermal ageing is driven by the annihilation of oxygen vacancies by water-derived species [6,7]. This process both destabilize the tetragonal phase and overstabilizes the monoclinic one [8]. Ageing kinetics are usually described by the Mehl–Avrami–Johnson equation [9] (eq. (1)).

$$\frac{V_m - V_0}{V_{max} - V_0} = 1 - exp(-(b.t)^n) \quad (1)$$

In this equation, the parameter b is thermally activated: $b = b_0 exp\left(\frac{-Q}{RT}\right)$.

V_m is the monoclinic phase content after ageing during a duration t at absolute temperature T, V_0 and V_{max} are the initial and saturation levels of monoclinic phase content, and R is the ideal gas constant. Q is the activation energy, typically around 100 kJ/mol. The value of the parameter n is typically between 0.3 and 3.5 [10].

Many factors such as different grain size, residual or applied stresses, density, content of Y_2O_3 in tetragonal phase, proportion of cubic phase, doping with more than one oxide, can influence the ageing process [4,11–17]. A few examples of these influences are given below. Jansen et al. reported that doping with ceria or decreasing the grain size strongly improved the ageing resistance of Y-TZP [18]. For instance, 3Y-TZP ceramics with grain size lower than 100 nm exhibit almost no ageing, whereas submicrometric 3Y-TZP ceramics may undergo severe degradation under the same conditions [19]. A critical value of grain size around 300 nm, below which ageing processes do not occur, was sometimes proposed

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https://doi.org/10.1016/j.actamat.2017.10.061
1359-6454/

[14], but later researches showed that this critical value has no physical basis [20] and may be dependent on other materials parameters [21]. Chevalier et al. reported that cubic grains are enriched by yttrium, which in turn leads to a decrease of the yttrium content in the neighboring tetragonal grains, making these Y-poor tetragonal grains more susceptible to transformation into monoclinic phase and thus affecting the ageing behavior of zirconia [12].

However, all the parameters mentioned above are interdependent, in particular through processing parameters: for instance, increasing the sintering time may increase both the grain size and the proportion of cubic phase [22], and the amount of cubic phase may also influence internal stresses. It is thus quite difficult to understand what parameters directly influence ageing [23]. Therefore, by carefully analyzing the microstructure and ageing behavior of several zirconia ceramics fabricated from the same powder, this paper seeks to isolate and quantify the influence of grain sizes, content of Y_2O_3 in tetragonal phase and amount of cubic phase on ageing kinetics parameters (Vmax, n, b_0, Q) of 3Y-TZP.

2. Materials and methods

2.1. Material preparation

The samples were fabricated starting from Y-TZP powders (TZ3YE, Tosoh, Japan). TZ3YE contains 3 mol% (5.2 wt%) yttria plus controlled alumina and silica doping. The zirconia green bodies used in the present study were prepared by slip casting. Firstly, the slurry were prepared by dispersing 150 g of TZ3YE powder in 37.5 g H_2O, using 2.25 g Darvan 821A as dispersant. Complete dispersion was achieved by ball milling with 150 g Y-TZP milling balls during 48 h. The resulting slurries were then cast in porous plaster molds and dried in air at 25 °C for 7 days. Finally the green bodies were sintered in a programmable electrical furnace, under different sintering conditions so as to obtain different microstructures: two materials were obtained by conventional sintering (samples 1430-2h and 1430-5h); two other materials were obtained by two step sintering (conditions chosen to obtain fully dense materials with small grain size [24]: TSS1 and TSS2 materials); finally some series of samples obtained using the TSS2 sintering conditions were submitted to different post-sintering thermal treatment in order to modify their microstructure (materials PS1150-100h, PS1150-200h, PS1430-20h, PS1500-2h, PS1550-20h). The exact sintering cycles used for all materials are summarized in Table 1. All samples (at least 6 per material) were shaped as 10 × 10 mm plates, polished, and annealed after polishing to remove any extra residual stress possibly induced by machining and polishing. The density of all samples was then measured by using Arthur's method, based on Archimede's principle.

2.2. Microstructural analyses: grain size, proportion of cubic phase, yttria content

SEM observations were conducted using a Zeiss Supra55 VP microscope. For each group of samples, grain size was obtained from micrographs taken on both side of 4 samples, using the linear intercept method, and multiplying by 1.56 the average linear intercept length of at least 500 grains to correct the fact that they were intercepted at random positions (not necessarily at their diameter) in accordance with the method developed by Mendelsohn et al. [25].

The average Y_2O_3 content in tetragonal phase and amount of cubic phase were obtained from XRD data (using a D8 Advance diffractometer, Bruker, Germany, with step size 0.02 deg and scan speed 1 s/step). To summarize, first a calibration curve of the amount of Y_2O_3 in the cubic and tetragonal phases versus the lattice parameters was established, from PDF files and previous work by other authors [26–28] (Fig. 1). Then XRD patterns were recorded (on one sample per group) over a large angular range (15–120 deg. 2θ) and analyzed using an iterative procedure based on Rietveld refinement. Rietveld refinement was conducted with Topas 4.0 software (Bruker), using a fundamental parameters approach; only scale factors, lattice parameters, crystallite size and systematic diffractometer-related angular errors were set free during the refinements. The outcomes of the first iteration of Rietveld refinement were mainly the approximate proportions of the different phases (cubic and tetragonal) and the lattice parameters of the tetragonal phase (in most materials cubic peaks were lost in the background or convoluted with some tetragonal peaks). The amount of yttria in the tetragonal phase was then calculated from the calibration curve. Since the total amount of Y_2O_3 is known (3 mol.%) from the composition of the powder, it was thus possible to determine the amount of yttria in the cubic phase (considering the approximate proportion of cubic phase) and thus its lattice parameter. The next iterations were conducted while setting the cubic phase lattice parameter as a constant (calculated from the amount of yttria in the cubic phase), which allowed evaluating more and more precise phases proportions and tetragonal phase lattice parameter, thus recalculating a more and more precise cubic phase lattice parameter. The iterations were continued until convergence (2–4 iterations generally allowed convergence to the 3rd digit for the proportion of the phases and to the 6th digit for the lattice parameters).

2.3. Assessment of ageing kinetics parameters

Ageing kinetics were evaluated by performing accelerated ageing tests on all samples in water vapour in an autoclave (Wolf Sanoclav), at temperatures ranging from 80 to 150 °C.

The amount of tetragonal to monoclinic phase transformation was measured at different time points by X-ray diffraction (XRD). XRD patterns were recorded in the 27–33° (2θ) range with a scan speed of 0.2 deg·min^{-1} and a step size of 0.05°, using a Bruker D8-advance diffractometer. The monoclinic ZrO_2 phase content (V_m) [29] was calculated as:

$$V_m = \frac{1.311X_m}{1 + 0.311X_m} \quad \text{(2-a)}$$

In which the value of X_m was determined directly from the diffraction diagrams using Garvie and Nicholson's equation [30]:

$$X_m = \frac{I_m^{-111} + I_m^{111}}{I_m^{-111} + I_m^{111} + I_t^{101}} \quad \text{(2-b)}$$

Where I_p^{hkl} is the area of the diffraction peak related to the (hkl) plane of phase p (m for monoclinic and t for tetragonal).

The hydrothermal ageing kinetics were rationalized by fitting the transformation curves with the Mehl–Avrami–Johnson laws [9] as explained in the introduction. For materials TSS1, TSS2, 1430-2h and 1430-5h, complete ageing kinetics were recorded at 140, 134, 110 and 85 °C (one sample per temperature); then the ageing parameters (V_0, V_m, b_0, Q, n) were obtained using a fitting procedure consisting in minimizing the total quadratic error between