



Enhanced reliability of yttria-stabilized zirconia for dental applications



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ABSTRACT

An increasing number of dental applications based on yttria-stabilized zirconia (3Y-TZP) have been developed in recent years as a result of the advances and versatility of dry-processing and soft machining at the pre-sintered state. Nonetheless, the long-term surface stability of these materials in humid environment is still a matter of concern and may limit its application. In this work, a simple method to prevent hydrothermal degradation on the zirconia surface is studied in detail. This method involves the infiltration of pre-sintered parts with optimized solutions containing Ce salts, leaving unchanged the other processing steps, allowing the diffusion of Ce during conventional sintering. Several pre-sintering conditions, solution concentrations and sintering temperatures were studied and characterized, obtaining working parameters for the production of zirconia parts with mechanical properties similar to standard 3Y-TZP and high resistance to hydrothermal aging. This optimal combination was obtained with the 1150 °C pre-sintering temperature, 50 wt.% solution and sintering at 1450 °C, leading to a superficial CeO₂ content of about 3 mol.%.

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1. Introduction

Tetragonal polycrystalline zirconia stabilized with 3 mol.% of yttria (3Y-TZP) is a biocompatible ceramic with good mechanical properties and a white translucent color, which makes it suitable as a structural material in biomedical implants and dentistry. Transformation toughening is the mechanism behind the mechanical behavior of 3Y-TZP. The tetragonal phase, stable above 1000 °C in pure zirconia, is retained to room temperature in a metastable state by adding a suitable amount of yttria, which acts as a stabilizer. This metastable phase is able to undergo stress-induced transformation thanks to the high stresses arising in front of a crack under external loading. The product is the stable monoclinic phase, which has a specific volume approximately 4.5% higher than the tetragonal phase. This local transformation has a displacive character and the increase in volume generates compressive stresses around the growing crack, hindering its propagation and thus increasing the fracture toughness of the material. The transformability of the tetragonal phase is the key parameter in Y-TZP: the stabilizer content should be high enough to avoid transformation during cooling from the sintering temperature, but low enough

to guarantee metastability in the tetragonal phase so that $t\text{-}m$ transformation can be activated by stress. With 3 mol.% of yttria, a fair combination of bending strength (higher than 1000 MPa) and fracture toughness (about 5 MPa \sqrt{m}) can be achieved [1].

1.1. Zirconia in arthroplasty and dentistry

Zirconia started to be employed in total hip arthroplasty in the late 80's, when it became an alternative to alumina for manufacturing femoral heads, since its higher strength and fracture toughness could offer more flexibility in terms of design and the possibility to implant less invasive prosthesis. In the last years, 3Y-TZP has also been used for the production of various dental devices (crowns, bridges, abutments, fixed partial dentures-FPDs and dental implants), where translucency and easy color modification for matching teeth shade represent big advantages for 3Y-TZP with respect to metal and metal–ceramic parts [2,3].

The production of 3Y-TZP femoral heads dropped drastically after finding that slight alterations in the processing steps could lead to failure of ceramic balls partly due to low-temperature degradation (LTD) [4]. This phenomenon, often referred to as hydrothermal degradation or aging, occurs in humid environments and at moderate temperatures (including human body temperature). LTD consists of the spontaneous and progressive formation of monoclinic phase under hydrothermal exposure, inducing surface roughening, localized stress, microcracking and often grain

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pull-out. The effects of LTD become evident when degraded parts are subjected to wear since large quantities of debris are produced and the wear rate dramatically increases with respect to non-degraded zirconia [5]. Even though in near fully-dense 3Y-TZP parts LTD may only interest a superficial layer of a few micrometers, in poorly sintered pieces with significant residual porosity, the bulk may be affected leading eventually to failure [6]. For these reasons, new composites of alumina/zirconia (zirconia-toughened alumina, ZTA) have been developed, which offer similar or improved mechanical properties and are barely affected by LTD.

However, 3Y-TZP is now attracting increasing interest in dentistry, thanks to the latest improvements in CAD/CAM processing and the possibility to color the visible parts by doping or by veneering to match the natural teeth shade [7,8]. It is also becoming clear that zirconia can be osseoconductive, so dental implants with tailored superficial porosity and roughness are being developed [9]. In these applications, a solid anchor of the implant to the bone is needed during the whole life of the patient. Due to the relatively high cyclic contact loading during mastication, crack nucleation and propagation by subcritical crack growth should be avoided, therefore the effect of the environment must be considered. In the current generation of 3Y-TZP, a small content of Al (less than 0.5%) is added in order to reduce the LTD kinetics [10], but the material still suffers from this phenomenon. Even though the effects of LTD on the flexural strength of dental zirconia under monotonic and cyclic loading seems to be limited and dependent on the aging time [11,12], this issue is still a matter of concern [13–15] and represents an impediment to the development of all-ceramic dental systems. Better solutions are therefore needed to overcome the problem, which is the subject of this work.

1.2. Ce doping in zirconia

The presence of vacancies in the ZrO_2 lattice generated by Y_2O_3 addition has been recognized as the key point for stabilization of the tetragonal phase at room temperature. During hydrothermal aging these vacancies may operate as hosting sites for the diffusion of water species from the surrounding environment, reducing their number and destabilizing the tetragonal crystal structure [16]. Other stabilizers can be employed instead of Yttrium. In the case of Cerium, due to the tetravalent character of Ce^{4+} ion that forms solid solution with zirconia, the vacancy mechanism is not operative, and therefore the stabilization of the tetragonal phase given by Ce^{4+} is imputable to the oversized dimension of this ion. Ce^{4+} forms a random substitutional solution into the Zr tetragonal lattice, but adopts a more symmetric 8-fold coordination due to the bigger size of Ce^{4+} with respect to Zr^{4+} , resulting in decreased tetragonality of the unit cell, and the stabilization of the tetragonal and cubic phases [17].

Ce-stabilized zirconia (Ce-TZP) is far more resistant to hydrothermal aging than 3Y-TZP [18]. However, both 10 Ce-TZP and 3Y-TZP have almost the same t - m transformation temperature T_0 , implying a similar driving force for transformation. Therefore, materials with the same driving force (as can be estimated in terms of T_0) can exhibit very different LTD kinetics [18,19]. The slower kinetics in Ce-TZP may be reasonably related to the lack of extrinsic vacancies when Ce is still in the initial oxidized state 4+. The first steps of degradation may thus be blocked and this could explain why aging is strongly retarded. Nevertheless, hydrothermal aging is still present in Ce-TZP, with very sluggish kinetics, so the explanation offered is not sufficient. Probably, the vacancies naturally present in zirconia lattice at room temperature or retained after sintering are responsible for inducing aging. The presence of a limited amount of Ce^{3+} ions is also not to be excluded [20]. Other authors state that ceria apparently segregates into grain boundary or produces a thin film of CeO_2 on the surface of the sam-

ple, preserving Y-TZP “active points” that would be responsible for initiating the aging phenomenon [21].

Cerium can be added to Y-TZP improving significantly the aging resistance. Nonetheless, it has been observed that the co-doping increases the grain size and somehow reduces the sintering capabilities of the material leaving some residual porosity [22]. Indeed, the development of new materials stabilized with both Ce and Y will be confronted to the challenge of finding the right balance between Ce and Y in order to optimize both mechanical and aging behaviors. Increasing Ce and Y contents may produce high hydrothermal aging resistance but low fracture toughness because of lower transformability [18] as it occurs with the sole Y doping when its content is increased over 3 mol.%. On the other side, doping with only high Ce concentration makes the material immune to LTD but also induces low hardness and strength.

Another possible strategy is to increase the Ce content only in the superficial region. Diffusion of ceria in the surface of zirconia was performed by heat-treating Y-TZP into ceria powder beds [23]. In a similar fashion, Marro et al. [24] have either pressed a layer of CeO_2 powder on the surface of dense 3Y-TZP or deposited a thin film of ceria on the surface, followed by annealing to allow diffusion into 3Y-TZP. As a result, hydrothermal aging was prevented on the Ce-rich face in both cases, without impairing surface mechanical properties with respect to the standard 3Y-TZP.

The objective of the present paper is precisely to study a method for adding ceria to 3Y-TZP, with higher concentrations in the surface, in order to improve its long-term surface stability. The method should easily adapt to dry processing of zirconia ceramic parts, commonly used to manufacture dental prosthesis. The latter starts from spray-dried granules of sub-micrometric powders with tailored characteristics, which are usually pressed isostatically in a deformable mold. The green piece is then pre-sintered in air, obtaining a porous machinable preform which can be soft machined via CAD/CAM before sintering in air between 1350 and 1600 °C [25]. Infiltration processing was chosen for this purpose since it can be easily adapted to dry-processing. By this method, the pre-sintered blank is infiltrated with a Ce solution after soft machining, leaving unchanged the other usual processing steps.

1.3. Infiltration processing

Infiltration processing has been employed to add alumina to zirconia by soaking pre-sintered cylinders into molten Al nitrate salts [26]. Infiltration with Ce acetate was adopted for coloring purposes, observing no change in mechanical properties up to concentration of 5 wt.% in water solutions [7]. Ce nitrate has been used for powder coating since this highly soluble salt distributes evenly once the solvent is evaporated, obtaining CeO_2 after calcination [27].

In Duh et al. [28], pellets of pressed zirconia, either mixed with CeO_2 or Y_2O_3 by the co-precipitation method, were infiltrated in the green state with Y nitrate and Ce nitrate solutions, respectively, and the properties of the resulting materials were compared, mainly in terms of fracture toughness and aging behavior. Unfortunately, no information was provided on the strength, the actual yttria and ceria contents, or their distribution in the final materials. Finally, it was shown in a previous communication that infiltration of dental posts in the pre-sintered state with Ce nitrate did not reduce the strength [29].

In the present work, we investigate processing conditions for the co-doping of 3Y-TZP with Ce by liquid infiltration with the objective of increasing significantly the aging resistance without affecting negatively mechanical properties. For doing so, we describe the influence of several Ce solution concentrations,

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