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Feature article

Fractography of self-glazed zirconia with improved reliability

Zhijian Shen[a,*], Leifeng Liu[a], Xiqing Xu[a], Jing Zhao[a], Mirva Eriksson[a], Yuan Zhong[a], Erik Adolfsson[b], Yihong Liu[c], Andraž Kocjan[d]

[a] Department of Materials and Environmental Chemistry, Arrhenius Laboratory, Stockholm University, S-106 91 Stockholm, Sweden
[b] Swerea IVF, Argongatan 30, S-431 53 Mölndal, Sweden
[c] Department of General Dentistry, Peking University School and Hospital of Stomatology, 100081 Beijing, China
[d] Department for Nanostructured Materials, Jožef Stefan Institute, Jamova 39, SI-1000 Ljubljana, Slovenia

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ABSTRACT

The fractography of a new grade of zirconia ceramics, known as self-glazed zirconia, was investigated. The as-sintered intact top surface was made with superior smoothness that mimicked the optical appearances of the natural teeth enamel. The beneath surface opposite to this was made hierarchically rough with microscopic pits of the size up to 60 μm together with grain-level roughness of about 2 μm. The three-point bending test of the samples made with the hierarchically rough surface being tensile one demonstrated an average bending strength of 1120 ± 70 MPa and a Weibull modulus of as high as 18 ascribed to the improved structural homogeneity. Surface topography was found the main origins of crack initiation leading to fracture. The observed unusually predominant transgranular fracture mode of submicron-sized grains disclosed a possible toughening mechanism of disassembling of mesocrystalline grains that differs significantly from the commonly quoted phase transformation toughening of this category of ceramics.

1. Introduction

Zirconia ceramics, commonly in the form of 3 mol-% Y_2O_3 partially stabilized ZrO_2 (3Y-TZP), are increasingly applied for fabrication of dental restorations due to their biocompatibility, mechanical reliability and aesthetic advantages [1–3]. Though zirconia ceramics are known as the toughest family of ceramics [4], clinical failures by fracture have been observed, particularly with the current tendency of adding increased amount of stabilizer in order to increase the content of cubic phase aiming for improving their optical translucency [5,6]. To solve these problems, materials with properties beyond those of ceramics used today must be developed. The general efforts to achieve this have so far mainly been focused on the development of new advanced ceramics and ceramic composites with a drastically reduced grain size [7–9], which has reminded the importance of powder processing in obtaining the required homogeneous microstructure and the key role of processing defects in determining the strength and reliability of ceramic materials [10,11].

Strength measurements are thus usually performed on samples with carefully prepared surfaces to exclude the artificial, extrinsic surface defects introduced by the post-process machining [12]. The strength of a ceramic product containing such extrinsic surface defects is known to be lower than that measured on test bars with carefully prepared surfaces. This means that the material's strength data reported in the literature or obtained from the properties data sheet provided by the manufactures in most of the cases represent only the maximum strength achievable but not that of the components made of such materials. The latter is however essential in predicating the reliability of the ceramic dental restorations having not-polished surfaces with customized geometries. In practice, extrinsic surface defects are further introduced by chairside grinding often required for restoring the demanded occlusion, which in turn would not only increase the surface roughness of the dental prostheses thus the wear of the opposite teeth but also the reliability of the dental prostheses themselves [13,14].

In this work the fracture behaviors of a new grade of self-glazed zirconia prepared by a precision additive 3D gel deposition approach based on hybrid gelation principle were investigated. Self-glazed zirconia was defined as a family of monolithic zirconia ceramics having a superior smooth surface opposite to another hierarchically rough surface both spontaneously formed during the net-shape forming process. While the former is made for imitating

* Corresponding author.
E-mail address: shen@mmk.su.se (Z. Shen).

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the function and optical appearance of natural tooth enamel, the latter is made for enhancing the bonding between ceramic prostheses and natural teeth or abutments above the dental implants [15]. With adjustable optical translucency and aesthetic behaviors the self-glazed zirconia family of ceramics fit particularly for model-free digital workflow of the manufacture of customized full-contour monolithic dental restorations by avoiding conventional manual work of grinding/polishing, veneering and glazing. In practice, their reliability would mainly be determined by the possible fracture initiated on the hierarchically rough surface under tension.

The performed work based on three-point bending test of the samples made with the hierarchically rough surface being tensile one followed by careful electron microscopic fractographic characterization of the fractured surfaces is aiming for disclosing the clues how can a combinational advantage of increased mechanical reliability and improved optical translucency be integrated into this new grade of zirconia ceramics. The fracture behaviors will be correlated to the processing defects with scales span from nanometers up to several tens micrometers, introduced during the entire manufacturing chain from powder synthesis to the sintering of bulk products.

2. Experimental

2.1. Preparation of the test samples

Self-glazed zirconia ceramic test samples were formed by a precision additive 3D gel deposition approach in the similar way as the production of customized self-glazed zirconia dental prostheses, so the samples contained one hierarchically rough surface opposite to the smooth self-glazed surface, both being spontaneously formed during a net-shape forming process based on hybrid gelation principle. All samples were pressure-less sintered in a muffle furnace at 1450 °C for 90 min in air to achieve a relative density above 99.9%. After that, the samples were furnace cooled down to the room temperature. The self-glazed zirconia is a product under development with the commercialization potential thus the processing details can not be disclosed here due to the conflict of interest.

2.2. Characterization of mechanical properties

The as-sintered hierarchically rough surface of 11 bending bars (1.5 × 2 × 30 mm) were evaluated by three-point bending test, using a fixture with a distance of 20 mm between the outer rollers in an universal testing machine (Zwick Z050, Zwick GmbH & Co. KG, Ulm, Germany) at a loading rate of 1 mm/min.

Instrument indentation test was carried out and load-displacement curves were obtained using a Fischerscope H100C nano hardness tester equipped with Vickers diamond indenter. The maximum load of 1 N was applied with the holding time of 10 s.

The hardness and fracture toughness of the sintered samples were measured by a Zwick/Roell ZHV indenter (Zwick/Roenhjll,Ulm, Germany), under a load of 10 kgf with a dwell time of 10 s. The hardness values were determined through the expression listed below [16],

$$H = 1.8544\frac{P}{d^2} \quad (1)$$

where, H is the hardness, P is the applied load, and d is the diagonal of the indentation.

The indentation fracture toughness was then calculated directly from the crack lengths using the equation given by Niihara et al. for Palmqvist cracks [17],

$$K_{IC} = 0.035\left(\frac{l}{a}\right)^{-1/2}\left(\frac{H}{E\Phi}\right)^{-2/5}\left(\frac{Ha^{1/2}}{\Phi}\right) \quad (2)$$

where, Φ is the constraint factor (≈3), E is the elastic modulus (here for zirconia ceramics it is measured to be 234 GPa through instrument indentation test), a is the half-diagonal of the Vickers indent, and l is the crack length measured from the indent edge.

2.3. Phase and microstructure characterization

Phase composition of the as-sintered and fractured surfaces of mechanically tested samples were characterized with X-ray powder diffraction using Panalytical Xpert PRO diffractometer (PANalytical, Almelo, Netherlands) in Bragg-Brentano geometry with Cu-K_α radiation ($\lambda = 1.5418$ Å). Measurements were made over a range of $20^\circ < 2\theta < 80^\circ$. The average crystallite size was determined by Scherrer equation [18]

$$D = k\lambda/B\cos\theta \quad (3)$$

Where, k is a constant rely on particle shape (0.89 for spherical particles), λ is the wave length of the X-ray ($\lambda = 0.154056$ nm for Cu Kα radiation), θ is the diffraction angle and B is the full width at half maximum (FWHM) of diffraction peak.

The microstructure was characterized by using a scanning electron microscope (SEM, JSM-7401F, JEOL, Tokyo, Japan). The as-sintered and fractured samples were washed by water and acetone in an ultrasonic bath before loaded into the SEM and the SEM observation was carried out on the surface without any coating. Accelerating voltages of 1.5 kV was applied in order to reduce the charging up of the samples. Besides, the cross-sections of a green body and an as-sintered sample were polished by Ar ion beam and SEM imaged at accelerating voltages of 8 kV under backscattered electron model to reveal the crystallographic orientation contrast. The grain size of the as-sintered samples was measured using the linear intercept method based on the SEM images (ASTM E0112-10) by the imaging software Smileview, with a three-dimensional correction factor of 1.2. FEI Titan transmission electron microscope (TEM) operating at 300 KV was used for TEM investigation of the zirconia starting powder and sintered bulks.

3. Results and discussion

3.1. Microscopic homogeneity

Fig. 1 shows a low magnification optical microscopic image and a high magnification SEM micrograph taken on the as sintered hierarchically rough surface of a tested sample. It appears that while the material is dense with rather homogeneous and fine grained microstructure, this surface reveals a hierarchically rough surface topography containing microscopic pits with size up to 60 μm and grain-level roughness of about 2 μm. The measured average grain size on this surface is 310 ± 90 nm.

Fig. 2 shows two backscattered SEM micrographs taken on Ar ion beam polished cross-section of a green body after burning-off organic binder and on a pressure-less sintered test sample, respectively. A homogeneous close packing of the starting nanoparticles achieved in the green stage is clearly demonstrated in Fig. 2(a). The low coordination number of uniformly distributed individual voids noticeable in this image would indicate a superior sinterablity of the obtained green body formed *via* such a precision additive 3D gel deposition approach. The fact that there are only fewer nano-sized residual pores visible on the Ar ion beam polished cross-section of the sintered test sample (see Fig. 2(b)) confirms that full densification is achieved by pressure-less sintering. An average grain size of 250 ± 70 nm is determined on the polished cross-section of the as-sintered sample, which is smaller than that observed on the as-sintered tensile surface. The enhanced surface atomic diffusion on free surfaces stimulated by optical radiation at high temperature