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Effect of grain size on the monoclinic transformation, hardness, roughness, and modulus of aged partially stabilized zirconia

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ABSTRACT

Objective. Low-temperature-degradation (LTD) has been reported to cause property changes in yttria-tetragonal zirconia polycrystals (Y-TZP). The current study measured monoclinic phase transformation of Y-TZP with different grain sizes and corresponding property changes due to artificial aging. Null hypothesis: the grain size of aged Y-TZP will not influence its transformation, roughness, hardness or modulus of elasticity.

Methods. Four groups of Y-TZP were examined with differing grain sizes ($n = 5$). The line intercept technique was used to determine grain sizes on SEM images (100,000×). Artificial aging was accomplished by autoclaving at 2 bar pressure for 5 h. X-ray diffraction (30 mA, 40 kV) was used to measure tetragonal to monoclinic transformation ($t \rightarrow m$). Surface roughness analysis was performed using a non-contact surface-profilometer. Nano-hardness and modulus of elasticity were measured using nano-indentation.

Results. SEM analyses showed different grain sizes for each sample group (0.350 μm, 0.372 μm, 0.428 μm, and 0.574 μm). The fraction of $t \rightarrow m$ transformation increased as grain size increased; furthermore, aging of zirconia caused increased roughness. Modulus and hardness after aging displayed no significant correlation or interaction with grain size.

Significance. Smaller grains caused less transformation, and aging caused increased roughness, but grain size did not influence the amount of increased surface roughness. Future studies are needed to determine the effects of grain size on the wear and fracture properties of dental zirconia.

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1. Introduction

Yttria-stabilized tetragonal zirconia polycrystal (Y-TZP) has become ubiquitous in prosthetic dentistry due to its high strength and biocompatibility. Y-TZP zirconia is stabilized in its less stable but more compact tetragonal crystal phase, however, thermal or mechanical stimuli will cause transformation into the more stable and less compact monoclinic crystal phase. This phase transformation imparts zirconia with its toughness; as cracks are formed in zirconia, their propagation initiates transformation of crystalline grains resulting in localized volumetric expansion and compression of the crack tip [1].

Phase transformation can also occur as the zirconia ages in a process known as low temperature degradation (LTD) [2]. The phase transformation and associated volumetric expansion that occurs in LTD can be detrimental to zirconia restorations. Volumetric expansion can cause internal stresses and micro-cracks, which could lead to premature failure of a zirconia structure [3]. Furthermore, the surface of aged, or transformed, zirconia has been shown to increase in roughness [4]. Increased surface roughness of dental zirconia could lead to higher wear rates of antagonist enamel in a zirconia dental restoration. Additionally, monoclinic transformation has been shown to diminish properties such as flexural strength, modulus of rupture [2,3,5], modulus of elasticity and hardness [6–10].

Recent trends in the fabrication of dental crowns and fixed partial dentures have shifted toward full contour zirconia crowns. In order to overcome the esthetic challenges associated with the opacity of monolithic zirconia, more translucent zirconia has been developed by decreasing its grain size [11]. Aside from the optical effects, decreasing grain size has been shown to stabilize the $t \rightarrow m$ transformation that occurs in LTD [2]. Reducing this $t \rightarrow m$ transformation should limit surface volumetric expansion and associated surface roughness. Additionally, less transformation may prevent degradation of the mechanical properties that has been reported for simulated LTD [6–10]. When the grains of yttria-stabilized zirconia go below a certain size (approximately 0.2 μm), however, the $t \rightarrow m$ transformation is no longer possible, which results in undesirable effects such as reduced fracture toughness [12].

In the current study, two brands of Y-TZP will be examined in their original composition (LAVA and LK2) and following modification to produce smaller grain sizes (LAVA Plus and LK1). The initial grain size of the zirconia was investigated to determine the effects of grain size on the $t \rightarrow m$ transformation due to artificial accelerated aging. Additionally, the associated changes in surface roughness, modulus of elasticity, and nano-hardness were measured before and after aging. The null hypotheses are: (1) grain size of the Y-TZP will not influence the amount of transformation or surface roughness after aging and (2) aging will not influence the modulus of elasticity or nano-hardness of the Y-TZP.

2. Materials and methods

2.1. *Zirconia characterization*

Y-TZP samples of various grain sizes were supplied by their manufacturer (LAVA and LAVA Plus; 3M ESPE, St. Paul, MN, USA/LK1 and LK2; LK Inter Corp, Taiwan) in the post-sintered polished state. A thorough study was performed by looking at samples that were supplied by two different manufactures with different proprietary stabilizers and chemical compositions to determine the effect of grain size. Furthermore, the samples were not independently polished, because the samples could only be statistically observed within the same manufacturer groups due to different proprietary chemical compositions. One sample from each group was thermally etched for 10 min at 1400 °C to identify grain boundaries [13]. Any changes in grain size that occurred during thermal etching could not be accounted for. Each etched sample was then sputter coated with gold/palladium and imaged in a high-resolution scanning electron microscope (SEM) (Quanta FEG 650; FEI, Hillsboro, OR, USA) at 100,000× to measure the grain sizes (Fig. 1A–D). The average grain-size was determined using the line-intercept technique on five micrographs for each sample. The circular-intercept technique was performed, using a 2 μm diameter circle, according to the Hillard Single-Circle Procedure [14] with a correction factor of 1.56 for polycrystalline ceramics according to Eq. (1) [15]:

$$G_{AVG} = 1.56 \left(\frac{\mathit{Circumference}}{n} \right) \tag{1}$$

2.2. *Artificial aging*

Artificial accelerated aging was accomplished using an autoclave device (Midmark M11 Ultraclave Automatic Sterilizer; Midmark Corporation, Versailles, OH, USA), programmed to run at 2 bar pressure for 5 h (2B5H) to induce severe aging, in accordance to the ISO standard 13356:2008 reference. Artificial aging due to autoclaving has been reported in the dental literature to sufficiently age dental zirconia and is viable for in vitro aging of zirconia [16]. Based on previously reported studies, this program simulates up to 15–20 years of actual aging [17,18].

2.3. *X-ray diffraction*

X-ray diffraction (XRD) is a method to measure the amount of monoclinic phase fraction present in stabilized zirconia. XRD was accomplished using a Siemens D 500 X-ray diffraction device (Siemens Corporation, Washington, D.C., USA) with Cu Kα radiation. Scans were performed at 40 kV, 30 mA, 0.02 degrees/step from 25° to 33°, and a dwell time of 12 s per step. Phase transformation was measured for each specimen ($n = 5$) prior to and following artificial aging to determine the amount of transformation. Representative plots are displayed in Fig. 2. The fraction of monoclinic phase can be calculated with the

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