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Research Paper

The influence of low-temperature degradation and cyclic loading on the fracture resistance of monolithic zirconia molar crowns

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ABSTRACT

The present study analyzed the kinetics of low-temperature degradation (LTD) in zirconia, and evaluated the influence of LTD and cyclic loading on the fracture resistance of monolithic zirconia molar crowns. Bar-shaped zirconia specimens were divided into nine groups and autoclaved at 134 °C for 0–200 h to induce LTD. The surface fraction and penetration depth of the monoclinic phase were examined using X-ray diffraction and scanning electron microscopy. Monolithic zirconia molar crowns were prepared for crown fracture testing. The crowns were autoclaved for 0–100 h ($n=6$) and cemented to dies. Six crown-die samples that were not autoclaved and six samples that were autoclaved for 100 h were subjected to cyclic loading with a load of 300 N for 240,000 cycles. All samples were tested in a load-to-failure test. The monoclinic fraction on the surface increased with autoclaving time and reached a plateau after 50 h. The depth of the monoclinic phase increased without reaching a plateau. The fracture load of the crowns significantly decreased from 5683 N (SD: 342) to 3975 N (SD: 194) after 100 h of autoclaving. Cyclic loading did not significantly affect the fracture resistance of the crowns in all cases. Kinetic analysis showed no linear correlation between the surface fraction and depth of the monoclinic phase after 50 h of autoclaving. Even though LTD increased the monoclinic phase, resulting in lower strength, the fracture resistance of the monolithic zirconia crowns was still sufficient to withstand the loading conditions in the molar regions.

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1. Introduction

The development of translucent tooth-colored zirconia enables the fabrication of restorations without using veneering porcelain (*i.e.* monolithic zirconia crown) (Johansson et al., 2014; Stober et al., 2014). Recently, it was demonstrated that monolithic zirconia crowns with a minimal occlusal thickness of 0.5 mm could withstand a bite force in the molar region because of the high fracture resistance (Nakamura et al., 2015). Since reducing the crown thickness contributes to the preservation of tooth substance, it can be a novel alternative dental prosthetic treatment with acceptable aesthetics.

Zirconia exhibits three crystal structures depending on the temperature. These crystal structures are the monoclinic, tetragonal and cubic phases (Chevalier et al., 2009; Scott, 1975). Pure zirconia without stabilizers has a tetragonal phase at a sintering temperature above 1170 °C. When cooling after sintering, a phase transformation occurs from tetragonal to monoclinic, which is accompanied by a volume expansion of around 4% that generates stress within the material. This causes fractures of the sintered material (Kelly and Ball 1986). Therefore, zirconia for dental prostheses is often stabilized with 3 mol% yttria (Denry and Kelly 2008). The stabilized zirconia consists of a metastable tetragonal phase. This metastable phase makes it possible to use bulk zirconia at room temperature. Also, the metastable tetragonal phase is resistant to crack propagation since it will be transformed to a monoclinic phase when exposed to mechanical stress. The phase transformation generates compressive stress because of the volume expansion in localized areas around micro-cracks, resulting in arrested crack propagation. This phenomenon is known as transformation toughening (Garvie et al., 1975). It is also known that the metastable tetragonal phase transforms into the monoclinic phase in a humid atmosphere without mechanical stress, beginning at the surface and entering the bulk of the material. This causes micro-cracks and a decrease in the strength of the material. This process is often referred to as low-temperature degradation (LTD) or aging (Chevalier et al., 2007). LTD is enhanced at temperatures of 200–300 °C (Chevalier et al., 1999), but can also occur *in vivo*. It has been reported that the zirconia femoral heads in hip joint prostheses can fracture or suffer from surface deterioration related to LTD (Chevalier et al., 2007; Haraguchi et al., 2001).

The influence of LTD on dental prostheses is not fully assessed (Lughi and Sergo, 2010). When the zirconia core is veneered with dental porcelain (*i.e.* zirconia-based crowns), zirconia is not directly exposed to the oral environment or to saliva. Thus, the influence of LTD could be limited. However, monolithic zirconia crowns will be directly exposed to saliva. Therefore, it is reasonable to assume that LTD might occur. Also, monolithic zirconia crowns will be directly and repeatedly loaded during mastication. This is known to affect the strength of all-ceramic crowns (Attia and Kern, 2004). Cyclic loading and LTD together may reduce the fracture resistance of monolithic zirconia crowns. To the best of the authors' knowledge, there are few available data on the strength of monolithic zirconia crowns.

The purpose of the present study was to analyze the kinetics of LTD in zirconia used for monolithic crowns, and to evaluate the influence of LTD and cyclic loading on the fracture resistance of monolithic zirconia crowns. The hypothesis was that the surface fraction and penetration depth of the monoclinic phase should increase as a result of LTD induced by autoclaving the samples. As a consequence of the increased monoclinic phase, the strength of the monolithic zirconia crown should decrease, especially when accompanied by cyclic loading, which was also the hypothesis of this study.

2. Materials and methods

2.1. Preparation of specimens

Sixty specimens of zirconia with dimensions of 17 × 7 × 1.8 mm were cut from zirconia blocks (Lava Plus Zirconia, 3M/ESPE St. Paul, MN, USA) using an Isomet 4000 (Buehler, Lake Bluff, IL, USA). The specimens were sintered at 1450 °C for 2 h (Lava Furnace 200, 3M/ESPE) according to the manufacturer's instructions. After sintering, the dimensions of the specimens were 13.5 × 5.5 × 1.5 mm. One side of each specimen was polished using 9, 6 and 1 μm diamond suspensions. Six specimens were allocated for grain size analysis and the others were used for analysis of the phase transformations. To analyze the grain size, the polished surfaces of the specimens were thermally etched for 30 min at 1400 °C. The average grain size was determined using the mean linear intercept technique with a scanning electron microscope (SEM; EM-3000, Topcon, Tokyo, Japan) according to a standard (ASTM Standard E112-13 2013). The density was measured using the Archimedes method.

2.2. Analysis of the crystalline phase transformation

Fifty-four of the prepared zirconia specimens were divided into nine groups ($n=6$). The specimens were immersed in distilled water and treated at 134 °C for 0, 10, 20, 30, 40, 50, 100, 150 and 200 h at 0.2 MPa in an autoclave (LSX-300, Tomy Seiko, Tokyo, Japan) to induce LTD. The temperature for autoclaving was determined according to ISO standard 13356:2008 (ISO13356, 2008). After autoclaving, the specimens were cut in the middle vertically in the direction of the long axis. One piece of the specimen was used for analysis with X-ray diffraction (XRD). The crystalline phase on the polished surface of the specimens was analyzed. The XRD data were collected with a θ–2θ diffractometer (X'Pert MPD, PANalytical, Tokyo, Japan) using CuKα radiation. Diffractograms were obtained from 27° to 33° at scan speed of 0.3°/min and a step size of 0.02°. The monoclinic phase fraction, *Xm*, was calculated using the Garvie and Nicholson (1972) method,

$$Xm = \frac{[Im(-111) + Im(111)]}{[Im(-111) + Im(111) + It(101)]},$$

where *It* and *Im* represent the integrated intensity of the tetragonal (101) and monoclinic (111) and (−111) peaks. The integrated intensity of each peak was calculated using HighScore Plus software (PANalytical). The monoclinic phase fraction is expressed as the percentage of tetragonal phase that was transformed to the monoclinic phase. Furthermore, in order to quantify the cubic phase content, the specimens without autoclaving were subjected to an additional XRD analysis ($n=3$). Diffractograms were obtained from 10° to 100° at scan speed of